TOPICS IN THE THEORY OF INHOMOGENEOUS MEDIA: COMPOSITE SUPERCONDUCTORS AND DIELECTRICS

DISSERTATION

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

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ABSTRACT

For composite superconductors, three topics are studied. First, a fully frustrated three-dimensional $XY$ model is studied using Monte Carlo calculations, finite-size scaling, and renormalization group methods. We find that the system has a continuous phase transition with critical temperature $T_c = 0.681 J/k_B$ and critical exponents $\alpha/\nu = 0.87 \pm 0.01$, $\nu/\nu = 0.82 \pm 0.01$, and $\nu = 0.72 \pm 0.07$. Second, the superconductor–insulator transition of a disordered 2D superconducting film as a function of the applied magnetic field is studied using quantum Monte Carlo calculations of the $(2+1)D$ $XY$ model. The magnetic phase factor $A_{ij}$ is assumed to have a mean of zero and a standard deviation of $\Delta A_{ij}$. The critical coupling constant $K_c$ and the universal conductivity $\sigma^*$ at $K_c$ are found for several values of $\Delta A_{ij}$. Three different phases, superconductor, Mott insulator, and Bose glass, are identified in the phase diagram of $1/K_c$ vs. $\Delta A_{ij}$. Third, the intermodulation coefficient of inhomogeneous high-$T_c$ cuprate superconductors with spatially varying gaps is calculated using an analogy to the description for an inhomogeneous dielectric with a nonzero cubic nonlinearity. Depending on the topology of the system, the intermodulation critical supercurrent density $J_{IMD}$ is enhanced, leading to a desirable material property.

For composite dielectrics, two topics are studied. First, a method to calculate the electric force acting on a sphere in two dielectric spheres immersed in a host...
with a different dielectric constant is described. The method uses a spectral representation, so the force is presented in a closed analytic form. The force between spheres approaches the dipole-dipole limit when the separation between spheres is very large. Then, the photonic band structures of metallic inverse opals above and below a plasma frequency $\omega_p$ is calculated. A plane wave expansion for the electric field $\mathbf{E}$ and the magnetic field $\mathbf{B}$ is used for the calculations. We obtain the same results using either field for $\omega > \omega_p$. We find that the plane wave method converges well for $\omega > \omega_p$, but not for $\omega < \omega_p$. 

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This is dedicated to my late grandmother and my late father, both passed away while I was pursuing a Ph. D.
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5.9 The radial component of the time-averaged force between two identical spheres of SrTiO$_3$ separated by $R$, plotted as a function of frequency for host materials consisting of silicone oil [(a) and (b)] and N$_2$ [(c) and (d)], with gap spacings $\delta = 0.10$ mm and $\delta = 0.30$ mm. The applied electric field is $E_0 = 71.3$ V/mm and $a = 3.15$ mm for all the cases. The electric field is parallel to the line between two spheres in (a) and (c), and perpendicular to that line in (b) and (d).  

5.10 The radial component of the time-averaged force between two identical spheres with $\epsilon_i$, plotted as a function of frequency $\omega/\omega_p$, where $\omega_p$ is the plasma frequency, for different $\ell_{\text{max}}$. It is assumed that $\epsilon_i$ follows the Drude model with $\omega_p \tau = 20$ and that the host material has $\epsilon_h = 1$. The gap spacing between two spheres is $\delta = 0.01$ mm. The applied electric field is $E_0 = 25.2$ V/mm and $a = 3.15$ mm. The electric field is parallel to the line between two spheres in (a) and perpendicular to that line in (b).  

6.1 Photonic band structures for a metallic inverse opal with void radius $R = 150$ nm and lattice constant $a = 500\sqrt{2}$ nm in the frequency range $\omega_p \leq \omega < 16\pi c/a$, where we use $\omega_p a/(2\pi c) = 7.694$. The number of plane waves used in this calculation is 537 and we expand the $E$ field of the modes.  

6.2 Same as Fig. 6.1, except $\omega a/(2\pi c) \leq 3$. These bands correspond to $\omega < \omega_p$.  

6.3 Same as Fig. 6.2, except that only a few ($\leq 10$) of the lowest eigenvalues are plotted for three different numbers of plane waves used: 339, 537, and 749, at frequencies such that $\omega < \omega_p$.  

6.4 Same as Fig. 6.1, except that $\omega_p < \omega < 6\pi c/a$ and $\omega_p a/c = 1$.  

6.5 Same as Fig. 6.4, except that $\omega a/(2\pi c) < 0.05$.  

6.6 Same as Fig. 6.5, except that $0.2\pi c/a < \omega < \omega_p$ and we expand the $B$ field of the modes.  

6.7 Same as Fig. 6.4, except $\omega_p a/(2\pi c) = 1$.  

6.8 Same as Fig. 6.7, except $\omega a/(2\pi c) < 0.5$.  

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6.9 Tight-binding results for $\omega < \omega_p$ with $a = 500\sqrt{2}$nm, $R = 150$nm, and $\omega_p a/c = 1$, using $\omega_{\text{int}} a/(2\pi c) = 0.1296$ for $\ell = 1$ in an infinite medium.
The horizontal dotted line represents the atomic level.

6.10 Tight-binding results for $\omega < \omega_p$ plotted together with those from the plane wave expansion for the $E$ field. The number of plane waves used in the plane wave expansion is 537.

6.11 Same as Fig. 6.10, except the plane wave expansion for the $B$ field.
CHAPTER 1

INTRODUCTION

The subject of this dissertation is categorized largely into two parts. One part is about composite superconductors and the other part is about composite dielectrics. The first part has topics on the continuous phase transition of a fully frustrated XY model in three dimensions, the intermodulation coefficient of an inhomogeneous superconductor, and the quantum Monte Carlo (QMC) study of a magnetic-field-driven two-dimensional (2D) superconductor–insulator (S-I) transition. The second part has topics on a method to calculate electrical forces acting on a sphere in an electrorheological fluid and the photonic band structures of metal inverse opals. The first part is quantum mechanics while the second part is classical ones. The intermodulation chapter plays the role of a bridge between quantum mechanics and classical ones. The QMC study of 2D S-I transition can also be thought of as an extension of the fully frustrated 3D XY model. The rest of this dissertation is organized as follows.

Chapter 2 studies a fully frustrated three-dimensional (3D) XY model on a simple cubic lattice. This model corresponds to a lattice of Josephson-coupled superconducting grains in an applied magnetic field $\mathbf{H} = (\Phi_0/a^2)(1/2,1/2,1/2)$, where $\Phi_0 = hc/2e$ is the flux quantum and $a$ is the lattice constant. We find the type of phase transition,
the value of the critical temperature, and the values of some critical exponents using several methods.

Chapter 3 will study the superconductor–insulator phase transition of a disordered 2D superconducting film as a function of the applied magnetic field, using quantum Monte Carlo calculations of the \((2 + 1)D\) \(XY\) model. We calculate the values of the critical coupling constant \(K_c\) and the universal conductivity \(\sigma^*\) for several values of \(\Delta A_{ij}\). From the results, we will discuss the transition from a superconductor to a Mott insulator and that from a Bose glass to the Mott insulator.

Chapter 4 describes a theory to describe how the inhomogeneity affects the intermodulation coefficient of high-\(T_c\) cuprate superconductors. We show that the continuum equations describing intermodulation in a superconducting layer with spatially varying properties are formally equivalent to those describing an inhomogeneous dielectric with a nonzero cubic nonlinearity. Using this formal analogy, we calculate the effect of inhomogeneity on the intermodulation coefficient in a high-\(T_c\) material, using several assumptions about the topology of the layer, and some simple analytical approximations to treat the nonlinearity.

Chapter 5 will introduce a new method to calculate electrical forces acting on a sphere in an electrorheological fluid. The method uses a spectral representation for the total electrostatic energy of the composite. The force is expressed as a certain gradient of this energy, which can be expressed in a closed analytic form rather than evaluated as a numerical derivative. The method is applicable even when both the spheres and the host have frequency-dependent dielectric functions and nonzero conductivities, provided the system is in the quasistatic regime. In principle, it includes all multipolar
contributions to the force, and it can be used to calculate multibody as well as pairwise forces.

Chapter 6 will calculate the photonic band structures of metallic inverse opals above and below a plasma frequency $\omega_p$, assuming a Drude dielectric function for the metallic component. For most of our calculations, we solve for the band structure using a plane wave expansion for either the electric field $\mathbf{E}$ or the magnetic field $\mathbf{B}$. We also use a single-cavity approach and a simple tight-binding approach for frequencies $\omega < \omega_p$.

Chapter 7 summarizes the results of this dissertation and gives some discussions.
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Composite Superconductors
CHAPTER 2

CONTINUOUS PHASE TRANSITION OF A FULLY FRUSTRATED \(XY\) MODEL IN THREE DIMENSIONS

2.1 Introduction

The classical \(XY\) model has been widely used for decades as a model for phase transitions in materials with interacting spins. The dimensionality \(n\) of the spins (\(n = 2\) for the \(XY\) model) is independent of the lattice dimensionality \(d\), which can be either 2 or 3 for physically relevant systems. The \(d = 2\) \(XY\) model undergoes the well-known Kosterlitz-Thouless (KT) transition [101], characterized by an unbinding of vortex-antivortex pairs at the KT transition temperature \(T_{KT}\). The KT transition is a continuous phase transition, but with unique critical properties [101]. The \(d = 3\) \(XY\) model exhibits a more conventional phase transition with well-known critical exponents [111, 65]. It is thought to describe many ferromagnetic materials with two-component spins. In addition, it describes phase transitions in which the “spins” actually represent the phases of a complex order parameter, such as the \(\lambda\) transition in He\(^4\) and superconductor-to-normal phase transition at zero applied magnetic field.

The frustrated classical \(XY\) model, in either \(d = 2\) or \(d = 3\), has a much wider range of phase diagrams than does the unfrustrated case just mentioned. In this
model, the coupling between spins is such that the ground state of the system cannot
minimize all the bond energies simultaneously. Interest in this model was greatly in-
tcreased when it was realized that this model described real systems, such as Josephson
junction arrays in an applied magnetic field. The first demonstration that the fully
frustrated $XY$ model undergoes a continuous phase transition in $d = 2$ was given
by Teitel and Jayaprakash [178], using Monte Carlo techniques. This work was later
extended to other values of the so-called frustration parameter $f$ [177], leading to an
extensive literature on 2D frustrated $XY$ model on various lattices and at different
values of $f$ [28, 64, 48].

The $d = 3$ frustrated $XY$ model has also been studied extensively, in part because
it is believed to describe flux line lattice (FLL) melting [124] under an applied mag-
netic field in high-$T_c$ superconductors. Hetzel et al. [77] used a uniformly frustrated
3D $XY$ model on a stacked triangular lattice to study the melting of an unpinned
Abrikosov lattice in a type-II superconductor. They showed convincingly that this
melting transition is first order, rather than continuous—a prediction subsequently
confirmed by experiment. Earlier work on a frustrated $XY$ model on a simple cubic
lattice, with magnetic field parallel to one of the lattice axes [155] found a continuous
phase transition. Li and Teitel [112, 113] used a uniformly frustrated $XY$ model
similar to that of Ref. [77] to calculate the properties of the vortex line liquid which
appears above the melting temperature, and to investigate the possibility of a further
phase transition between an entangled and a disentangled vortex line liquid above
the FLL melting transition. Chen and Teitel [31] later extended this work to the
more realistic case of uniaxially anisotropic couplings; they suggested that there was
another phase transition temperature $T_{cz}$ above the FLL melting temperature $T_m$.,
where superconducting coherence parallel to the applied magnetic field would vanish. Chin et al. [34] have, however, suggested that this apparent existence of a transition to a disentangled vortex liquid is a result of finite system sizes and simulation times. More recently, the uniformly frustrated 3D XY model was studied by Monte Carlo methods on a simple cubic lattice with a magnetic field parallel to the [111] direction [81]. For this choice of field direction, the simple cubic lattice behaves as a stack of 2D triangular lattices with $ABCABC\cdots$ stacking. It was found that this system, similar to that studied in Ref. [77], exhibits a clear first-order FLL melting transition. Nguyen and Sudbø [129] considered a uniformly frustrated anisotropic Villain model (an approximation to the uniformly frustrated XY model) to study the phase diagram of a uniaxially anisotropic high-$T_c$ superconductor as a function of the applied magnetic field and temperature. They found two phase transitions: the lower-temperature one is FLL melting, while that at higher temperature involves the destruction of the phase coherence in the direction of the applied magnetic field.

In this chapter, we study phase transitions in the fully frustrated XY model in $d = 3$, using primarily MC simulations. This model is of interest, in part, because it may be relevant to FLL melting in the high-$T_c$ materials. In addition, because of great advances in microfabrication techniques, it is now possible to make microscale or nanoscale arrays of Josephson superconducting grains in three dimensions. Such an array should, in a suitable applied magnetic field, be describable by a frustrated XY model, at least to a first approximation. It has also recently been suggested that a fully frustrated XY model might also be realized by an assembly of cold atoms on a suitably constructed optical lattice [144].
The fully frustrated $XY$ model is characterized by the frustration vector $\mathbf{f} = (1/2, 1/2, 1/2)$, as further defined below. Such a model has been previously studied by Diep et al. [47], who found that, in contrast to the $\mathbf{f} = (1/3, 1/3, 1/3)$ case studied in Ref. [81], there was a continuous phase transition. We extend the work of Ref. [47] by calculating the critical behavior of the helicity modulus, equivalent to the spin-wave stiffness constant (or to the superfluid density in a superconductor). We also carry out a more extensive finite-size scaling analysis than done by those workers, thus obtaining more accurate information about the critical behavior. Our results do, however, confirm that the phase transition is continuous.

The remainder of this chapter is organized as follows. In Sec. 2.2, we present the formalism for calculating the thermodynamic properties and the critical exponents of our model. In Sec. 2.3, we give the results of our Monte Carlo calculations, finite-size scaling methods, and renormalization group methods. Section 2.4 presents a summary and discussions.

2.2 Formalism

We now describe the model Hamiltonian on which our calculations are carried out. For convenience, we present this Hamiltonian as it applies to a simple cubic lattice of superconducting grains in the presence of an applied external magnetic field $\mathbf{H}$, though the model is not limited to this application, of course. Then the frustrated $XY$ model in $d = 3$ is described by the Hamiltonian

$$\mathcal{H} = -\sum_{\langle ij \rangle} J_{ij} \cos(\phi_i - \phi_j - A_{ij}).$$

(2.1)

Here $\phi_i$ is the phase of the superconducting order parameter on the $i$th site, $A_{ij} = \int_i^j \mathbf{A} \cdot d\mathbf{l}$, $\Phi_0 = hc/2e$ is the flux quantum, $\mathbf{A}$ is the vector potential, $J_{ij}$ is the
coupling constant between the $i$th site and $j$th site, and $h_{ij}$ represents a sum over all distinct pairs of nearest-neighbor sites on a simple cubic lattice. We assume a constant coupling between each nearest-neighbor pair of sites, so $J_{ij} = J$ and $J > 0$; we neglect the possible dependence of $J$ on the applied magnetic field $\mathbf{H}$ and the temperature $T$. We also assume weak screening as in Ref. [81]. Thus the local magnetic field $\mathbf{B}$ is approximated by the applied magnetic field $\mathbf{H}$.

The $x$ component of the frustration vector $\mathbf{f} = (f_x, f_y, f_z)$ is defined by

$$\sum_p^{(x)} A_{ij} = 2\pi f_x,$$ (2.2)

where the sum is taken along the sides of a plaquette on the $yz$ plane of the lattice; analogous definitions hold for $f_y$ and $f_z$. If the simple cubic lattice has lattice constant $a$, $f_i = B_ia^2/\Phi_0$ represents the flux through a single plaquette perpendicular to the $i$th axis, in units of one flux quantum.

One can also define the vortex number (or vorticity) of each square plaquette. For example, the vorticity of a plaquette lying in the $yz$ plane is defined by

$$n_x = f_x + \frac{1}{2\pi} \sum_p^{(x)} (\phi_i - \phi_j - A_{ij}),$$ (2.3)

where the sum runs counterclockwise around the perimeter of the plaquette, viewed from the positive $x$ direction, and the phase differences are chosen so that $0 \leq \phi_i < 2\pi$. $n_y$ and $n_z$ are defined analogously.

In order to study possible phase transitions within this model, we have calculated several quantities. One of these is the specific heat $C_V$ per site, given by

$$C_V = \frac{\langle \mathcal{H}^2 \rangle - \langle \mathcal{H} \rangle^2}{Nk_B T^2},$$ (2.4)

where $N$ is the total number of sites in the lattice, $\mathcal{H}$ is the Hamiltonian in Eq. (2.1), and $\langle \cdots \rangle$ denotes an average within the canonical ensemble. A first-order
phase transition is generally indicated by a \( \delta \)-function-like anomaly in \( C_V \), while a continuous phase transition is signaled by lattice-size-dependent divergence in the \( C_V \).

To study the vortex lattice melting, we calculate a suitable vorticity density-density correlation function. Specifically, we first introduce the Fourier transform of the vorticity density \( n_i(k) = \sum_R n_i(R) \exp(i k \cdot R) \), where \( n_i(R) \) represents the vorticity of a plaquette centered at \( R \) and oriented perpendicular to the \( i \)th axis. We then calculate some of the correlation functions

\[
g_{ij}(r) = \frac{1}{N^2} \sum_{R,R'} \langle n_i(R)n_j(R') \rangle,
\]

where the sum is carried out over all \( R \) and \( R' \) such that \( R' - R = r \). In our actual simulations, we use periodic boundary conditions in all three directions. In practice, it is more convenient to compute the Fourier transform of \( g_{ij}(r) \). This Fourier transform is known as the vortex structure factor, and is given by \( S_{ij}(k) = \sum_r g_{ij}(r)e^{ik \cdot r} \). Because of the periodic boundary conditions, \( S_{ij}(k) \) is defined only for \( k = [2\pi/(Na)](m_1, m_2, m_3) \), where \( m_i \) are integers, and the computational cell is assumed to contain \( N = N_x^3 \) sites. We obtain \( g_{ij}(r) \) by first computing \( S_{ij}(k) \) and then Fourier-transforming back into real space.

The phase transition in this model is best characterized by the **helicity modulus tensor** \( \gamma_{\alpha\beta} \) [56]. \( \gamma_{\alpha\beta} \) measures the stiffness of the phase \( \phi \) against an external twist. It is defined as the second derivative of the free energy with respect to an infinitesimal phase twist [135]

\[
\gamma_{\alpha\beta} = \frac{1}{N} \left. \frac{\partial^2 F}{\partial \delta_\alpha \partial \delta_\beta} \right|_{\delta=0}.
\]

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\( \gamma_{\alpha\beta} \) is conveniently calculated by adding a fictitious vector potential \( A' \) to the Hamiltonian in Eq. (2.1), as

\[
\gamma_{\alpha\beta} = \frac{1}{N} \left. \frac{\partial^2 F}{\partial A'_i \partial A'_j} \right|_{A'=0}, \tag{2.7}
\]
in the presence of periodic boundary conditions on the phases. The resulting diagonal components are readily evaluated, with the result

\[
\gamma_{\alpha\alpha} = \frac{1}{N} \left\langle \sum_{\langle ij \rangle} J_{ij} \cos(\phi_i - \phi_j - A_{ij})(\hat{e}_{ij} \cdot \hat{e}_\alpha)^2 \right\rangle
- \frac{1}{Nk_B T} \left\langle \left[ \sum_{\langle ij \rangle} J_{ij} \sin(\phi_i - \phi_j - A_{ij})(\hat{e}_{ij} \cdot \hat{e}_\alpha) \right]^2 \right\rangle
+ \frac{1}{Nk_B T} \left. \left. \left\langle \sum_{\langle ij \rangle} J_{ij} \sin(\phi_i - \phi_j - A_{ij})(\hat{e}_{ij} \cdot \hat{e}_\alpha) \right\rangle^2 \right\rangle. \tag{2.8}
\]

Here \( \hat{e}_{ij} \) is the unit vector from the \( i \)th site to the \( j \)th site, and \( \hat{e}_\alpha \) is the unit vector in the \( \alpha \) direction. Since the helicity modulus \( \gamma_{\alpha\alpha} \) is proportional to the \( \alpha \) component of the superfluid density, it follows that when \( \gamma_{\alpha\alpha} > 0 \), there is nonzero phase coherence in the \( \alpha \) direction, and when \( \gamma_{\alpha\alpha} \to 0 \), the phase coherence is lost. Therefore, the superconductor-to-normal phase transition occurs at the temperature at which \( \gamma_{\alpha\alpha} \to 0 \).

The correlation time \( \tau \) is a measure of how long it takes for the system to lose its memory of its previous state. \( \tau \) can be obtained by calculating a time-displaced autocorrelation function [125]. The time-displaced autocorrelation function \( \chi_E(t) \) of the energy, for example, is defined by

\[
\chi_E(t) = \int \left[ E(t') - \langle E \rangle \right] \left[ E(t' + t) - \langle E \rangle \right] dt'
= \int \left[ E(t') E(t' + t) - \langle E \rangle^2 \right] dt', \tag{2.9}
\]
where \( t \) and \( t' \) are two different Monte Carlo times.
Equation (2.9) can be expressed more conveniently when we have a set of measurements of the energy $E(t)$ from $t = 0$ (after equilibration) up to some maximum time $t_{\text{max}}$. In this case, Eq. (2.9) becomes

$$\chi E(t) = \frac{1}{t_{\text{max}} - t} \sum_{t' = 0}^{t_{\text{max}} - t} E(t') E(t' + t) - \frac{1}{(t_{\text{max}} - t)^2} \sum_{t' = 0}^{t_{\text{max}} - t} E(t') \sum_{t'' = 0}^{t_{\text{max}} - t} E(t' + t').$$  (2.10)

Since the autocorrelation function is expected to fall off exponentially at long times as

$$\chi E(t) \sim e^{-t/\tau},$$  (2.11)

the correlation time $\tau$ can be calculated from

$$\int_0^\infty \frac{\chi E(t)}{\chi E(0)} dt = \int_0^\infty e^{-t/\tau} dt = \tau.$$  (2.12)

This expression for $\tau$ is also called integrated correlation time. MC measurements will be statistically independent only if they are separated by intervals of $\sim 2\tau$ or more. In addition, the correlation time $\tau$ is related to the dynamic exponent $z$ by

$$\tau \sim \xi^z \sim L^z,$$  (2.13)

where $\xi$ is the correlation length (equal to $L$ at the $T_c(L)$). The dynamic exponent measures the extent of the critical slowing down. The smaller $z$ is, the more accurate are the numerical measurements.

If there is a continuous phase transition, various quantities should exhibit singular behavior near the transition temperature $T_c$. If we define a reduced temperature by

$$t = \frac{T - T_c}{T_c},$$  (2.14)
then in the thermodynamic limit the correlation length $\xi$, the specific heat per site $C_V$, and the helicity modulus $\gamma$ near $T_c$ are expected to vary as

$$\xi \sim |t|^{-\nu},$$  \hspace{1cm} (2.15)

$$C_V \sim |t|^{-\alpha},$$  \hspace{1cm} (2.16)

$$\gamma \sim |t|^v,$$  \hspace{1cm} (2.17)

where $\nu$, $\alpha$, and $v$ are critical exponents. If $T_c$ is known, the simulation data can be fitted to the expected asymptotic form to obtain values of the critical exponents. But since $T_c$ is typically not known in advance, it is usually more accurate to use a different method. One such method is finite-size scaling, which extracts values for the critical exponents by investigating how measurements depend on the size $L$ of the system [126]. This procedure is carried out by expressing a quantity of interest in terms of the correlation length and then introducing a new dimensionless function, known as a \textit{scaling function}. For example, $C_V$ and $\gamma$ are expressed as

$$C_V(L, t) = L^{a/\nu} \tilde{C}_V(L^{1/\nu} t),$$  \hspace{1cm} (2.18)

$$\gamma(L, t) = L^{-v/\nu} \tilde{\gamma}(L^{1/\nu} t),$$  \hspace{1cm} (2.19)

where $L = N_x a$ is the linear system dimension. Since the scaling functions $\tilde{C}_V$ and $\tilde{\gamma}$ should depend on a single variable, we can make all the data for each system size $L$ fall on the same curve by appropriately adjusting the values of the critical exponents and $T_c$. When this happens, we assume that we have the correct values for these quantities.

Another method is the phenomenological renormalization group (PRG) [8, 133, 111]. To calculate the critical behavior of $\gamma$, for example, we consider two different
system sizes $L$ and $L'$ and introduce the ratio

$$P_\gamma(L, L', t) \equiv \frac{\gamma(L, t)}{\gamma(L', t)}. \tag{2.20}$$

From Eq. (2.19) this ratio becomes $P_\gamma = (L/L')^{-v/\nu}$ when $t = 0$. Therefore, if one plots two different curves of $P_\gamma(L, L', t)$ versus $t$ with the same ratio of $L/L'$, the temperature at which they intersect is $T_c$, and the value of $P_\gamma$ at that temperature yields the ratio $v/\nu$.

The critical temperature $T_c$, or its inverse value $K_c = J/(k_BT_c)$, and the critical exponent $\nu$ can also be determined from Binder’s fourth-order cumulant $U_L$ [20, 19, 21] defined by

$$U_L = 1 - \frac{\langle s^4 \rangle}{3\langle s^2 \rangle^2}, \tag{2.21}$$

where $s = (1/N)\sqrt{\langle \sum_i^N \cos \phi_i \rangle^2 + \langle \sum_i^N \sin \phi_i \rangle^2}$. The scaling form for the Binder’s cumulant is $U_L = \tilde{U}(L^{1/\nu}t)$, without any prefactor. Hence, $U_L$ can be Taylor expanded about $T_c$ as

$$U_L = U_0 + U_1 L^{1/\nu} \left(1 - \frac{T}{T_c}\right) + \cdots$$

$$= U_0 + U_1 L^{1/\nu} \left(1 - \frac{K_c}{K}\right) + \cdots. \tag{2.22}$$

If we plot $U_L$ for several values of $L$ as a function of temperature or its inverse value, they will intersect at the critical temperature $T_c$. To obtain the exponent $\nu$, we can calculate $(dU_L/dK)_{K=K_c}$, where $K = J/(k_BT)$. From Eq. (2.22), we find that

$$\frac{dU_L}{dK}\bigg|_{K_c} = \frac{U_1}{K_c} L^{1/\nu}. \tag{2.23}$$

Hence, the ratio of the two slopes for different values of $L$ gives $\nu$. 

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2.3 Monte Carlo Calculations

To carry out our Monte Carlo calculations, we used the standard Metropolis algorithm with periodic boundary conditions in all three directions. We started with a random phase configuration at temperature $T = 1.0J/k_B$, then cooled down to $T = 0.4J/k_B$ in steps of $0.01J/k_B$, except near $T_c$, where we decreased the temperature in steps of $0.005J/k_B$. At each $T$, we took 50000 Monte Carlo steps per site through the entire lattice for equilibration, and then calculated the expectation values of the quantities of interest by averaging over an additional 20000 MC steps. We also used the final configuration of the previous $T$ as the starting one of the current $T$. Near $T_c$, the system undergoes critical slowing down; so we increased the number of MC steps up to $5 \times 10^5$ for equilibration and $2 \times 10^5$ for averaging.

Instead of considering continuous angles between 0 and $2\pi$ for the phases $\phi_i$ of the order parameter on each site, we used the 360-state clock model, which allows angles of $0^\circ$, $1^\circ$, $2^\circ$, ..., $359^\circ$. This simplification should have no effect on our results, since it is known that there is no distinction between the continuum and the discrete results for the $n$-state clock model when $n > 20$ [179].

To calculate the phase factors $A_{ij}$, we use the gauge $\mathbf{A} = (\Phi_0/a^2)(f_y \hat{x} + f_z \hat{y} + f_x \hat{z})$, where the frustration is $f = f_x \hat{x} + f_y \hat{y} + f_z \hat{z}$. Thus, for example, the phase factors $A_{ij}^{(z)}$ arising from the field component parallel to $z$ all vanish except for bonds in the $y$ direction; for these bonds, and $x = na$, $A_{ij}^{(z)}$ is given by

$$A_{ij}^{(z)} = 2\pi nf_z. \quad (2.24)$$

The phase factors $A_{ij}^{(x)}$ and $A_{ij}^{(y)}$ are given by analogous expressions. For $f = (1/2, 1/2, 1/2)$ and periodic boundary conditions in all three directions, all the phase factors
are equal to either 0 or \( \pi \), and thus all the couplings are of the form \( \pm J \cos(\phi_i - \phi_j) \). This choice of gauge automatically satisfies the condition given in Eq. (2.2).

We turn now to our numerical results. The values of the correlation time \( \tau \) for several lattice sizes are given in Table 2.1, at the estimated transition temperature \( T_c(L) \) for the fully frustrated \( XY \) model \([f = (1/2, 1/2, 1/2)]\) of size \( L = N_x a \). \( \tau \) clearly increases monotonically with increasing lattice size. From the fits to data in Fig. 2.1, we get the dynamic exponent \( z = 2.23 \pm 0.14 \) in our system.

<table>
<thead>
<tr>
<th>( L )</th>
<th>( \tau(t_{MC}) )</th>
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<tr>
<td>4</td>
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<td>6</td>
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<td>14</td>
<td>164</td>
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<td>16</td>
<td>352</td>
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Table 2.1: Calculated correlation time \( \tau \) for several lattice sizes \( L \), evaluated at \( T_c(L) \). \( \tau \) is measured in the units of MC steps per site.

The internal energy per site \( U \equiv E/N \), expressed in units of the coupling constant \( J \), is shown in Fig. 2.2. It is quite size-dependent for \( L < 10 \) but quickly converges for \( L > 10 \). The sharp drop in \( U \) near \( T = 0.7J/k_B \) suggests but obviously does not prove the occurrence of a phase transition near that temperature.

The calculated specific heat per site \( C_V \) is shown in Fig. 2.3. There is a clear peak near \( T \sim 0.7J/k_B \) which becomes sharper with increasing \( L \), suggesting a continuous phase transition. This behavior is similar to that of the 2D fully frustrated \( XY \) model as in Ref. [178] and that of the ordered simple cubic lattice with \( f = (0, 0, 1/2) \), as
Figure 2.1: Log-log plot of the data in Table 2.1, corresponding to a fit of $\tau$ to the function $\tau = AL^z$. The slope of the fitting line to the data yields the dynamic exponent $z = 2.23 \pm 0.14$.

Figure 2.2: Internal energy per site as a function of temperature $T$ for several lattice sizes, as indicated. Note that $T$ decreases with increasing distance along the horizontal axis.
in Ref. [155]. As in those two examples, the finite magnitude of \( C_V \) at its peak is a result of the finite system size; otherwise, \( C_V \) would diverge at \( T_c \) in the infinite system.

![Figure 2.3](image)

Figure 2.3: The specific heat per site \( C_V \) as a function of temperature for several lattice sizes. The lines are cubic spline fits to the data.

Next, we turn to the behavior of the helicity modulus tensor. Since \( \gamma_{xx} = \gamma_{yy} = \gamma_{zz} \) for this isotropic system, we calculated the average \( \gamma = (\gamma_{xx} + \gamma_{yy} + \gamma_{zz})/3 \) in order to improve the statistics. \( \gamma(T) \) increases with decreasing \( T \), and shows a fairly clear drop to near zero near \( T = 0.7J/k_B \) when \( L > 10 \), as can be seen in Fig. 2.4. For \( L = 10 \) or smaller, \( \gamma(T) \) shows a broad transition region. A more accurate value for \( T_c \) will be given further below.

To estimate the statistical errors in our calculation of \( \gamma(T) \), we used the jackknife method [127], which is carried out as follows. For each \( T \), we made \( n \) independent
numerical measurements of \( \gamma(T) \), with measurements separated by at least two correlation times. From these \( n \) measurements, we calculated a value \( \gamma(T) \) for the helicity modulus. Next, we removed the first measurement from this set of \( n \) measurements to calculate the helicity modulus \( \gamma_1 \) with the remaining \( n - 1 \) measurements. To calculate \( \gamma_2 \) we restored the first measurement to the set and removed the second measurement, and so on. Thus, \( \gamma_i \) is calculated with the \( i \)th measurement removed from the set. The error estimate for \( \gamma \) is given by

\[
\sigma_\gamma = \sqrt{n \sum_{i=1}^{n} (\gamma_i - \gamma)^2}.
\]

The error bars from this method are shown in Fig. 2.4, but they are smaller than the symbol sizes.

In Fig. 2.5, we show the evolution of the internal energy per site \( U \) as a function of MC time at the transition temperature \( T_c = 0.681J/k_B \) for a lattice size \( 12 \times 12 \times 19 \).
12. The three intensity plots for each \( a \) and \( b \) show the density-density correlation functions \( g_{zz}(x, y, L_z/2) \), \( g_{xx}(L_x/2, y, z) \), and \( g_{yy}(x, L_y/2, z) \) (denoted by \( xy \), \( yz \), and \( zx \), respectively) of the vortices at two different times as indicated in the energy evolution curve. Each intensity plot is an average of one correlation time \( \tau = 148 \) MC steps per site through the entire lattice. The two times correspond to energies slightly below and slightly above the average value and they are separated by a very large MC time. In contrast to the results of Ref. \[81\], there is no indication of a vortex lattice phase in these plots; instead, both seem to show vortex liquid phases (although there are partial latticelike formations, especially on the \( zx \) plane in window \( a \)). This behavior suggests (as does the diverging specific heat peak in Fig. 2.3) that the phase transition at \( f = (1/2, 1/2, 1/2) \) is continuous, rather than first order as at \( f = (1/3, 1/3, 1/3) \). However, at very low temperatures \( (T = 0.01J/k_B) \), the correlation functions show a clearer indication of an ordered phase, as shown in Fig. 2.6. Specifically, we see evidence of a checkerboard lattice in all three directions, but especially in the \( yz \) windows.

This ordering is clearer if we look at the vortex structure factor \( S_{zz}(k) \) of the lattice. Figure 2.7 shows this structure factor for \( k \) parallel to the magnetic field, at several temperatures, including the two shown in Figs. 2.5 and 2.6. Here \( k_{||} = |k| \) and \( k = [2\pi/(N_xa)](m, m, m) \), where \( m = 0, 1, ..., N_x - 1 \). At low temperatures, the vortex structure factors at \( k_{||} = \sqrt{3}\pi/a \) are nonzero, but they do not increase monotonically with decreasing temperature. Instead, they increase down to \( T = 0.30J/k_B \), then decrease down to \( T = 0.01J/k_B \). We believe that this behavior may represent some kind of “polycrystalline” domain structure of the vortex at low temperatures. As shown below, the system has several eigenmodes, and the exact admixture of these
Figure 2.5: Internal energy per site for a lattice of size $12 \times 12 \times 12$ as a function of MC time, at the transition temperature $T_c = 0.681 J/k_B$. The intensity plots $a$ and $b$ represent the vorticity density-density correlation functions $g_{zz}(x, y, L_z/2)$, $g_{xx}(L_x/2, y, z)$, and $g_{yy}(x, L_y/2, z)$ denoted by $xy$, $yz$, and $zx$, respectively, at the times $a$ and $b$ in the energy evolution curve. A lighter color represents a larger value of the correlation function.

eigenmodes may change as the temperature varies. The insets to Fig. 2.7 are the vorticity density-density correlation functions $g_{zz}(x, y, L_z/2)$ at two different temperatures $T = 0.30 J/k_B$ and $T = 0.681 J/k_B$. We can see a clear checkerboard pattern at $T = 0.30 J/k_B$. The vortex structure factor appears to go to zero near the same temperature as $T_c$ where the helicity modulus vanishes, although we did not collect enough numerical data to determine the temperature dependence of the structure factor peak near $T_c$.

In Fig. 2.8, we show the probability distribution $P(U)$ of the internal energy per site $U$ for several lattice sizes at $T_c$. For each lattice size, we see only a single peak, which sharpens with increasing lattice size. This result is also in contrast to that of Ref. [81], where the single peak splits into two peaks as the lattice size increases. This
persistent single-peak behavior suggests a continuous phase transition, in contrast to the first-order phase transition found in Ref. [81].

The data shown in Fig. 2.3 can be used in a standard way to obtain an estimate of the critical exponent ratio $\alpha/\nu$. From Eqs. (2.15) and (2.16), the maximum value of $C_V$ for a lattice of edge $L$ is

$$C_V^{\text{max}} \sim \xi^{\alpha/\nu} \sim L^{\alpha/\nu}. \quad (2.26)$$

In Fig. 2.9, we plot $\log C_V^{\text{max}}$ versus $\log L$ at $T = T_c$. The data is well described by a straight line. The slope of this fitted straight line gives $\alpha/\nu = 0.87 \pm 0.01$. The present result is in contrast to the linear relation between $C_V$ and $\log L$, corresponding to $\alpha = 0$ (logarithmic divergence) seen in the fully frustrated 2D $XY$ model as in Ref. [178].
As we did for the specific heat $C_V$, we see from Eqs. (2.15) and (2.17), or just from Eq. (2.19), that the helicity modulus $\gamma$ depends on the lattice size $L$ as

$$\gamma \sim \xi^{-v/\nu} \sim L^{-v/\nu}.$$  \hfill (2.27)

In Fig. 2.10, we plot $\log \gamma$ versus $\log L$ at two different temperatures $T = 0.681J/k_B$ and $T = 0.682J/k_B$. From the fits to the data, we get $v/\nu = 0.79 \pm 0.02$ at $T = 0.681J/k_B$ and $v/\nu = 0.83\pm0.02$ at $T = 0.682J/k_B$. The error bars from the jackknife method are also shown, but they are smaller than the symbol sizes. The value of $v/\nu$ is very sensitive to small changes in the assumed $T_c$. So we need a more reliable method to calculate $v/\nu$. In Fig. 2.11, we show the results of a PRG study of the helicity modulus $\gamma(L,t)$. As described near Eq. (2.20), we plot $P_z(L,L',t)$ for the fixed ratio $L/L' = 2$, but for three different pairs of sizes $L$ and $L'$. From the intersection of
Figure 2.8: MC probability distribution \( P(U) \) for the internal energy per site \( U \) at the transition temperature \( T_c = 0.681 J/k_B \) for several lattice sizes.

Figure 2.9: Log-log plot of the maximum height \( C_V \) of the specific heat per site versus linear size \( L \) of the lattice. The points are MC data; the full curve is the best-fit line. The slope of the fit line gives \( \alpha/\nu = 0.87 \pm 0.01 \).
these three $P_\gamma(L, L', t)$ curves as a function of $t$, we find $T_c = 0.681 \pm 0.001J/k_B$ and $v/\nu = 0.82 \pm 0.01$.

Figure 2.10: Log-log plot of the helicity modulus $\gamma$ versus linear size $L$ of the lattice. The points are MC data at two different temperatures $T = 0.681J/k_B$ and $T = 0.682J/k_B$; the full curves are the best-fit lines. The slopes of the fit lines give $v/\nu = 0.79 \pm 0.02$ at $T = 0.681J/k_B$ and $v/\nu = 0.83 \pm 0.02$ at $T = 0.682J/k_B$. The error bars from the jackknife method are smaller than the symbol sizes.

The previous two methods give only the ratios of two critical exponents. In order to find all three critical exponents, we need a third method to extract $\nu$. For this, we use another renormalization group transformation [128]. We first calculate $U(T)$ for a $16 \times 16 \times 16$ lattice. Next, we reduce each linear lattice dimension by a factor of 2, by combining a cube of eight adjacent sites into one; this is known as a *blocking scheme* of the real-space renormalization group method. This factor 2 corresponds to the scaling factor $b$. The phase of the merged eight sites is assigned by a simple
Figure 2.11: PRG method to extract the transition temperature $T_c$ and the critical exponent $\nu/\nu$ of the helicity modulus $\gamma$. All three curves show $\gamma(L, t)/\gamma(L', t)$ for the same ratio of $L/L' = 2$. The $x$ and $y$ coordinates of the intersection of the three curves yield the values of $T_c$ and $(L/L')^{-\nu/\nu}$.

additive rule, that is, it is taken as

$$\phi_b = \arctan \left( \frac{\sum_i \sin \phi_i}{\sum_i \cos \phi_i} \right),$$  \hspace{1cm} (2.28)

where $-\pi \leq \phi_b \leq \pi$. We add $2\pi$ when $\phi_b < 0$ so that $\phi_b$ satisfies $0 \leq \phi_b < 2\pi$. The correlation length $\xi'$ of the blocked lattice is also reduced by the scaling factor $b$, i.e.,

$$\xi' = \frac{\xi}{b}$$ \hspace{1cm} (2.29)

when $\xi'$ is measured in terms of the lattice constant. But since the correlation length should diverge at $T_c$, it follows that

$$\xi(T_c) = \xi'(T_c).$$ \hspace{1cm} (2.30)

To use this scheme to obtain $\nu$, we also calculate the internal energy per site $U'$ of the rescaled $8 \times 8 \times 8$ lattice as a function of temperature $T$. This rescaled lattice
should behave very similarly to an $8 \times 8 \times 8$ lattice at a different temperature $T'$; so

$$U'(T) = U(T').$$  \hfill (2.31)

If we rewrite $T'$ in terms of $T$, we obtain

$$T' = U^{-1}[U'(T)],$$  \hfill (2.32)

where $U^{-1}$ is the functional inverse of the function $U$. This relation between $T$ and $T'$ is the desired renormalization group transformation. As in Eq. (2.15), the rescaled correlation length $\xi'$ can be expressed in terms of the reduced temperature $t'$ as

$$\xi' \sim |t'|^{-\nu}. \hfill (2.33)$$

From Eqs. (2.15), (2.29), and (2.33), we obtain

$$\left( \frac{t}{t'} \right)^{-\nu} = b. \hfill (2.34)$$

Therefore, knowledge of the renormalization group transformation taking the system from $T$ to $T'$ gives the value of $\nu$.

Since Eqs. (2.29) and (2.33) are the asymptotic forms near $T_c$, we can linearize the renormalization group transformation using a Taylor series expansion near $T_c$ to obtain

$$T' - T_c = (T - T_c) \left. \frac{dT'}{dT} \right|_{T_c}, \hfill (2.35)$$

or equivalently

$$\frac{t'}{t} = \left. \frac{dT'}{dT} \right|_{T_c}. \hfill (2.36)$$

Inserting this result into Eq. (2.34) yields

$$\nu = \frac{\log b}{\log \left. \frac{dT'}{dT} \right|_{T_c}}. \hfill (2.37)$$
In Fig. 2.12, we show simulation results for the rescaled $8 \times 8 \times 8$ lattice and for a separate unrescaled lattice of the same size. If we fit the two sets of points near the critical temperature $T_c$ to two straight lines, the ratio of the slopes yields the value of $(dT'/dT)_{T_c}$, according to Eq. (2.36). We can then get $\nu$ from Eq. (2.37). From the plot shown in Fig. 2.12, we obtain $\nu = 0.72 \pm 0.07$. Since we already know $\alpha/\nu$ and $v/\nu$, we can use this value of $\nu$ to obtain $\alpha = 0.63 \pm 0.07$ and $v = 0.59 \pm 0.07$.

![Figure 2.12: RGT method to extract the transition temperature $T_c$ and the critical exponent $\nu$ of the correlation length. The ratio of the slopes of the two fitting lines at $T_c$ gives the value of $\nu$, according to Eq. (2.37).](image)

All of our numerical data appears to be consistent with a single phase transition at $T_c = 0.681J/k_B$. By contrast, there are two separate phase transitions in 2D fully frustrated XY model on a square or triangular lattice, as discussed in Refs. [138, 139] and [100]. One of these is a KT transition while the other is in the Ising universality class. Although the two transition temperatures are close to each other,
the Ising transition temperature is slightly higher than that of the KT transition. Reference [100] explains the sequence of these phase transitions in terms of the loss of phase coupling across a domain wall. Although our results are consistent with a single phase transition, we do not have sufficient data to rule out the possibility that there could be two separate phase transitions in our model.

To gain further insight into our numerical results, we have also used the mean-field approximation developed by Shih and Stroud [156] to find the relevant eigenmodes near the phase transition at $T_c$. Near $T_c$, the linearized mean-field equations are

$$
\eta_i - \frac{\beta}{2} \sum_j J_{ij} e^{iA_{ij}} \eta_j = 0,
$$

(2.38)

where $\eta_i \equiv \langle e^{i\phi_i} \rangle$ and $\beta = 1/k_B T$. If we assume $J_{ij} = J$ and use $T' = k_B T / J$, Eq. (2.38) becomes

$$
\eta_i - \frac{1}{2T'} \sum_j e^{iA_{ij}} \eta_j = 0.
$$

(2.39)

In the mean-field approximation, $T_c$ is the highest value of $T$ such that Eq. (2.39) has a nontrivial solution. With our frustration $\Gamma = (1/2, 1/2, 1/2)$, the Hamiltonian is periodic with a unit cell of $2 \times 2 \times 2$, which implies that Eq. (2.39) is a set of eight coupled homogeneous linear equations. $T_c$ is given by the condition that the determinant of the matrix of coefficients should vanish. This requirement leads to two values of $T' = \pm \sqrt{3}/2 \sim 0.866$; each is fourfold degenerate. Thus the transition temperature in the mean-field approximation is $T_c^{MF} = 0.866 J/k_B$. The four degenerate eigenmodes corresponding to this eigenvalue are $(0, -1, 1, \sqrt{3}, 0, 0, 0, 1)$, $(1, 0, -\sqrt{3}, -1, 0, 0, 1, 0)$, $(-1, \sqrt{3}, 0, -1, 0, 1, 0, 0)$, and $(-\sqrt{3}, 1, 1, 0, 1, 0, 0, 0)$; these modes are shown in Fig. 2.13. Of course, any linear combination of these modes would also be an eigenmode. Since the four eigenmodes are degenerate, they must be
related by some discrete symmetry operations of the lattice. Thus, we might expect that there are several degenerate ground states. Indeed, our simulations at very low temperatures \( T = 0.01J/k_B \) do suggest that the system can readily fluctuate among several different states at such temperatures.

![Diagram of degenerate eigenmodes](image)

Figure 2.13: Four degenerate eigenmodes for the \( 2 \times 2 \times 2 \) unit cell of the ordered state with \( \mathbf{f} = (1/2, 1/2, 1/2) \), corresponding to the mean-field transition temperature \( T_c^{MF} = \sqrt{3}/2 \ J/k_B \). The numbers represent the order parameters \( \eta_i \) of the mode, as indicated in Eq. (2.39). The phases reside on the nodes of the lattice.

### 2.4 Discussion

We have investigated the phase transition in a fully frustrated 3D \( XY \) model on a simple cubic lattice, corresponding to an applied magnetic field \( \mathbf{H} = (\Phi_0/a^2)(1/2, 1/2, 1/2) \). In contrast to the case \( \mathbf{f} = (1/3, 1/3, 1/3) \) as in Ref. [81], we see a continuous
phase transition. We also extract the critical exponent ratios \( \alpha/\nu = 0.87 \pm 0.01 \), \( v/\nu = 0.82 \pm 0.01 \), and \( \nu \) itself, using a variety of numerical techniques. We get \( \alpha = 0.63 \pm 0.07 \), \( v = 0.59 \pm 0.07 \), and \( \nu = 0.72 \pm 0.07 \). Our \( T_c = 0.681 \pm 0.001 J/k_B \) is very close to that in Ref. [47], but they used a “crossover temperature” \( T_{co} \) to calculate \( \alpha \) and \( \nu \). As a result, they have two values for each \( \alpha \) and \( \nu \). Our \( v \) is never calculated before.

It is of interest to compare these values with those of other models. In the isotropic (unfrustrated) 3D XY model, \( \nu \sim 0.66 - 0.67 \) [111, 64, 84, 52, 154, 129, 149, 131, 103, 182], \( \alpha \approx 0 \) [64, 84, 149] (\( \alpha = -0.017 \) in Ref. [154]), and \( v \approx \nu \) [111, 64, 149, 129]. For the anisotropic but unfrustrated XY model in \( d = 3 \), Ref. [130] reported that \( \alpha \sim -0.007 \). In the weakly frustrated XY model in \( d = 3 \) (\( f = (0,0,f) \), with \( f \leq 1/12 \)), it has been reported that \( \nu \approx 1.5 \) [130]. Reference [90] found that \( \nu = 2.2 \pm 0.4 \) in a random-coupling 3D XY model with free boundary conditions and \( f = (0,0,1/(2\pi)) \), while Ref. [91] found that \( \nu = 1.1 \pm 0.2 \) in a random-coupling 3D XY model with periodic boundary conditions and \( f = (0,0,1/4) \). Our value of \( \nu \) satisfies the usual trend that \( \nu \) becomes larger than 0.66 – 0.67 when the magnetic field is nonzero. However, our results appear not to satisfy the two hyperscaling laws \( \alpha = 2 - d\nu \) and \( v = (d - 2)\nu \). Possibly the reason is that at this phase transition, two order parameters go to zero: the helicity modulus, and a discrete, Ising-like order parameter related to the amplitude of the structure factor at a characteristic wave vector.

The \( T_c \) we obtain from the PRG method for \( \gamma \) agrees very well with that found from the RGT method, and quite well also with that estimated from \( C_V(T) \). It is, however, about 20% lower than that obtained by the mean-field approximation. This
deviation is not a surprise, because the Monte Carlo $T_c$ has been shown to be lower than that found by mean-field theory in other frustrated $XY$ systems [155].

In order to avoid the problem of critical slowing down, instead of just increasing the number of Monte Carlo steps, one might attempt to use the cluster flipping algorithm, as first proposed by Swendsen and Wang [170], and later by Wolff [189], and many others [49, 132, 87, 88, 92, 27, 116, 115]. This algorithm is very successful in reducing critical slowing down for unfrustrated models such as the $O(n)\sigma$ model and the Potts model. But when the system is frustrated, especially fully frustrated, a mere extension of the cluster algorithm does not reduce the critical slowing down, and may even *increase* the correlation time [28]. The reason is that the cluster percolation temperature $T_p$ is much larger than $T_c$ in the frustrated system, rendering the cluster flipping trivial at $T_c$ because the percolating cluster takes up almost the entire system. Therefore, the cluster should be generated in such a way that $T_p$ is closer to $T_c$ [28, 27]. Critical slowing down might also be reduced if the standard Metropolis algorithm is combined with the cluster algorithm, resulting in a hybrid algorithm [131, 143, 103].

The renormalization group transformation method is based on certain assumptions which may lead to systematic errors [128]. Specifically, it assumes that the blocked system has a typical phase configuration of another 3D $XY$ model on a simple cubic lattice with $L' = L/b$, i. e., that they appear with the correct Boltzmann probabilities as the original states. This assumption is not exactly correct, and contributes to some systematic error, which cannot be easily estimated. In the present chapter, we have tried to estimate these errors using the jackknife method. The renormalization group method is also affected by finite-size effects, because the original system has a different size than does the rescaled system. To optimize the benefits of this method, therefore,
we should ideally run simulations on as large a system as possible. We should also do an extra simulation on a system whose size is the same as that of the rescaled system, and compare the two results.

We also comment briefly on the difference between our work and that of Diep et al. [47]. These authors did not compute the helicity modulus, nor did they comment on the connection between the model and a superconducting array in a magnetic field. In addition, because of the several techniques described above, we are able to get more accurate values of the ratio $\alpha/\nu$, as well as values for $\nu/\nu$ and of $\nu$ itself.

We also briefly discuss the possibility that the phase transition at $T_c$ might actually be two separate phase transitions. In the 2D fully frustrated $XY$ model, as already mentioned, there are indeed two separate phase transitions: a KT transition at a lower temperature, followed by an Ising-like transition at a slightly higher temperature [138, 139, 100] between a state in which the vortices are ordered in a checkerboard pattern and a disordered vortex state. In the present case, the transition at $T_c$ also involves the nearly simultaneous disappearance of 3D $XY$ order (signaled by the vanishing of the helicity modulus) and a discrete order parameter (indicated by the vanishing of the vortex structure factor). Once again, in 3D, the discrete order is characterized by a checkerboard configuration of the vortex state below $T_c$ (see Figs. 2.5–2.7), although in this case the discrete order parameter is described by four degenerate modes, as determined by the mean-field solution, rather than two as in the 2D fully frustrated $XY$ model [35].

Although the discrete and $XY$ order appear to vanish at the same temperature in 3D, and there is no evidence of two separate phase transitions, we believe that our numerical results are not sufficient to conclusively rule out two separate phase
transitions. In particular, we have not carried out careful numerical studies of the critical behavior of the discrete order parameter. It would be of interest to carry out further numerical studies, especially of the discrete order parameter, to answer this question definitively.

To summarize, we have studied the phase transition in the fully frustrated $XY$ model, using Monte Carlo simulations in conjunction with two types of real-space renormalization group approaches. We find, in agreement with previous work, that the phase transition is continuous, and we obtain accurate values of the critical exponents $\alpha/\nu$ and $\nu/\nu$, and a slightly less accurate value for $\nu$ itself. The phase transition could, in principle, be probed experimentally in a suitable 3D lattice of coupled superconducting grains, and possibly also in an assembly of cold atoms in an optical lattice.
CHAPTER 3

QUANTUM MONTE CARLO STUDY OF A MAGNETIC-FIELD-DRIVEN 2D SUPERCONDUCTOR–INSULATOR TRANSITION

3.1 Introduction

The superconductor–insulator transition of thin 2D superconducting films has been extensively studied both theoretically [29, 30, 55, 102, 148, 162, 86, 9, 117, 183, 75, 153, 109, 108, 134, 26, 160] and experimentally [73, 151, 118, 120, 192] for many years. The theoretical work can be largely categorized into two groups; one uses a random chemical potential as a disorder while the other uses a magnetic field as a disorder. Most of previous work belong to the former [29, 102, 148, 162, 86, 9, 117, 183, 109, 108, 160] whereas only a few belong to the latter [30, 134, 118, 120].

The present work is motivated primarily by several experiments in which an S-I transition is observed in a 2D material as a function of applied transverse magnetic field. Such experiments have been reported in thin films of superconducting materials. They have also been carried out in some of the most anisotropic cuprate high-$T_c$ superconductors; in such materials, individual copper oxide layers may conceivably behave like thin superconducting films if they are well enough decoupled from the other layers [74, 136, 63, 89, 137, 60, 119, 70, 59, 14, 165, 13, 166, 7]. In both cases,
the films seem to undergo a transition from S to I with increasing magnetic field. Furthermore, the transition appears to be controlled mainly by the film resistance $R$. Experiments suggest that, in contrast to some predictions, $R$ does not have a universal value at the S-I transition. In view of these experiments, it seems useful to construct a simple model which contains disorder and also shows a field-driven transition. In the present chapter we present such a model, and analyze its properties by a combination of numerical methods and scaling assumptions.

Before describing our own approach, we briefly review some of the previous theoretical work in this area. An early numerical calculation was carried out by Cha et al. [29] at zero magnetic field ($B = 0$). These workers calculated both analytically and numerically the zero-temperature ($T = 0$) universal conductivity $\sigma^*$ of the 2D boson-Hubbard model without disorder at the S-I transition. They found, using numerical MC simulations of a (2+1)D $XY$ model, that $\sigma^* = (0.285 \pm 0.02)\sigma_Q$, where $\sigma_Q = (2e)^2/h$ is the quantum conductance. This result is close to the value obtained from an analytic large-$N$ expansion. They further studied this model under an applied transverse magnetic field using MC simulations, and found that $\sigma^*$ was increased [30].

Fisher et al. [55] studied the phase diagrams and phase transitions of bosons with short-range repulsive interactions under periodic and random potentials at zero temperature. They found that bosons in periodic potentials at zero temperature showed two different phases, superfluid and Mott insulating phases, and that the dynamic exponent $z$ was exactly equal to the spatial dimension $d$ and the constrictions of the correlation length exponent $\nu$ and the order parameter correlation exponent $\eta$ were $\nu \geq 2/d$ and $\eta \leq 2 - d$ at zero-temperature superfluid–insulator transition.
They also found that a “Bose glass” phase existed in the presence of disorder and that the transition to a superfluid occurred from the Bose glass phase, not directly from the Mott insulator, in the presence of disorder.

Most of previous work used QMC simulations [102, 148, 162, 9, 117, 183, 75, 153, 109, 108, 26]. Some used advanced QMC techniques, such as a QMC algorithm based on the exact duality transformation of the boson Hubbard model [75] and a worm algorithm [108]. Others used a stochastic series expansion method [75, 160] and an exact diagonalization method [134]. Analytically, a coarse-graining approximation [86] was used other than the large-$N$ expansion method [29].

Some used a 2D hard core boson model [148, 117, 75, 153, 134] while others used a 2D soft core boson model. Some studied the superconductor–insulator phase transition at zero temperature [162, 86, 117, 134, 160, 151], some studied the superconductor–Bose-glass phase transition [148, 9], and others studies both [102, 183, 109, 108]. Smakov and Sorensen [160] studied the superconductor–insulator transition at finite temperature $T$. Capriotti et al. [26] studied a reentrant superconducting-to-normal phase transition using a resistively shunted 2D Josephson junction array with normal Ohmic shunt resistors as the source of dissipation. This reentrant superconducting-to-normal phase transition persisted for moderate dissipation strength. They also found that the superconducting phase was always stabilized beyond a critical dissipation strength at sufficiently low temperatures. Hébert et al. [75] studied the phase transitions between superfluid, checkerboard, and striped solid, using two parameters of disorder, the near ($V_1$) and next near ($V_2$) neighbor repulsion, instead of a single parameter for the random chemical potential. They found that the superfluid to striped solid transition at half filling and the transition from the superfluid to the
striped supersolid phase away from half filling were first order while the transition from the striped supersolid phase to the striped solid phase was second order. Schmid et al. [153] studied the first order transition between a checkerboard solid and a superfluid phase at a finite temperature. They also found an unusual reentrant behavior with ordering upon increasing temperature. As an effort to make the model more realistic, short- and long-range Coulomb interactions were included [162, 183] or the amplitude fluctuations were included in the order parameter in addition to the phase fluctuations [86].

At the zero-temperature superconductor–insulator transition, the dynamic exponent, the correlation length exponent, and the universal conductivity were found to be \( z = 0.5 \pm 0.1 \), \( \nu = 2.2 \pm 0.2 \), and \( \sigma_c = (1.2 \pm 0.2)\sigma_Q \), respectively, from QMC simulations [117]. The universal conductivities were also found to be \( \sigma^* = (\pi/8)\sigma_Q \) from the coarse-graining approximation [86] and \( \sigma^* = (0.45 \pm 0.05)\sigma_Q \) at finite temperature from the stochastic series expansion with a geometric worm algorithm [160], where conductivity scaled with \( \omega/T \) at small frequencies and low temperatures. With the short-range Coulomb interactions, the universal conductivity was found to be \( \sigma^* = (0.14 \pm 0.03)\sigma_Q \) [162, 183]. With the long-range Coulomb interactions, it was found to be \( \sigma^* = (0.55 \pm 0.1)\sigma_Q \) [162] or \( \sigma^* = (0.55 \pm 0.06)\sigma_Q \) [183].

In a magnetic field, Nishiyama [134] found that the 2D hard core boson model exhibited a field-tuned localization transition at a certain critical magnetic field and that the critical DC conductivity was substantially larger than that at zero magnetic field. He claimed that the critical conductivity was not universal but increased with the magnetic field. Experimentally, Sambandamurthy et al. [151] found the resistivity followed a power law dependence on the magnetic field in the superconducting as well
as insulating phases using thin films of amorphous InO near the superconductor–insulator transition.

At the zero-temperature superconductor–Bose-glass transition, the dynamic exponent and the universal conductivity were found to be \( z = 1.95 \pm 0.25 \) and \( \sigma^* = (0.17 \pm 0.01)\sigma_Q \), respectively [148]. Batrouni et al. [9] found \( \sigma^* = (0.45 \pm 0.07)\sigma_Q \) from the QMC calculations and \( \sigma^* = (0.47 \pm 0.08)\sigma_Q \) from the current-current correlation functions.

At the intermediate strength of disorder, the dynamic exponent and the correlation length exponent were found to be \( z = 1.35 \pm 0.05 \) and \( \nu = 0.67 \pm 0.03 \), respectively [109]. Lee et al. [109] also found that a Mott insulator to a superfluid transition occurred in the weak disorder regime and that a Bose glass to the superfluid transition occurred in the strong disorder regime. This was studied further with the behavior of the quasiparticle energy gap near the quantum phase transition by Lee and Cha [108]. They found that the gap vanished sharply at the transition for a weak disorder, implying a direct Mott insulator to superfluid transition, and that this feature disappeared for a strong disorder, supporting the intervention of Bose glass phase.

A \( d \)-dimensional quantum mechanical system can be mapped onto a \((d+1)\)-dimensional classical system with the imaginary time as an extra dimension [161]. This is because calculating the thermodynamic variables of the quantum system is equivalent to calculating the transition amplitudes of the classical system when they evolve in the imaginary time, of which interval is fixed by the given temperature. The net transition amplitude between two states of the system can be obtained by the summation over amplitudes of all possible paths between them according to Feynman [54]. These paths are the states of the system at each intermediate time step. Therefore the
path-integral description of the quantum system can be seen through the statistical mechanics of the classical system with a fictitious temperature that measures zero-point fluctuations in the quantum system.

In order for a boson system to have the superconductor–insulator transition, the system must have an on-site repulsive interaction, or a charging energy. Otherwise all bosons will have Bose-Einstein condensation at zero temperature. This charging energy induces zero-point fluctuations of the phases and disorders the system. On the other hand, the Josephson coupling, or $XY$ coupling favors coherent ordering of the phases, which causes the onset of superconductivity. Therefore the competition between the charging energy and $XY$ coupling is responsible for the superconductor–insulator transition. In addition, if a disorder is added to the system, the system will undergo a different phase than a Mott insulator depending on the strength of the disorder. That phase is known to be a Bose glass phase.

In this chapter, we study the zero-temperature quantum phase transitions of 2D superconducting films in an applied magnetic field. We begin with a boson Hubbard model, and use path-integral Monte Carlo calculations of the $(2 + 1)$D $XY$ model and finite-size scaling method to obtain the critical coupling constants and universal conductivities at the transition. We also use a positional disorder method and the renormalized coupling constant to investigate the behavior of the system in the fully random case. The formalism of our system is given in Sec. 3.2 and the numerical results are given in Sec. 3.3. We summarize this chapter and provide some discussions in Sec. 3.4.
3.2 Formalism

3.2.1 Model Hamiltonian

Our goal is to examine the superconductor–insulator transition in a disordered 2D system in a magnetic field at very low temperature $T$. Thus, a useful model for this transition will include three features: (i) a competition between a Coulomb energy and an energy describing the hopping of Cooper pairs; (ii) disorder; and (iii) a magnetic field. While there are a wide range of models which could incorporate these features, we chose to consider a model Hamiltonian appropriate to a 2D Josephson junction array:

$$H_0 = U \sum_j n_j^2 - J \sum_{\langle ij \rangle} \cos(\theta_i - \theta_j - A_{ij}).$$

(3.1)

Here $n_j$ is the operator representing the number of Cooper pairs on a site $j$, $J$ is the Josephson energy coupling sites $i$ and $j$, $A_{ij} = (2\pi/\Phi_0) \int_i^j \mathbf{A} \cdot d\mathbf{l}$ is a magnetic phase factor, $\Phi_0 = hc/2e$ is the flux quantum, and $\mathbf{A}$ is the vector potential. In this picture, each site can be thought of as a superconducting grain.

For calculational convenience, we choose to take the sites $j$ to lie on a regular 2D lattice (a square lattice in our calculations), with Josephson coupling only between nearest neighbors. Thus, the disorder in our model is incorporated via the magnetic phase factors $A_{ij}$, as explained further below. Our Hamiltonian is identical to that of Cha et al. [29] except that we consider the special case that the chemical potential $\mu_i$ for Cooper pairs on the $i$th grain is an integer, and we choose the $A_{ij}$’s to be random.

The first term in Eq. (3.1) is the charging energy. We consider only a diagonal charging energy and also assume all grains to be of the same size, so that $U$ is independent of $j$. Since the charging energy $E_{C,j}$ of a grain carrying charge $Q_j$ with
capacitance \( C \) is \( E_{Cj} = Q_j^2/(2C) \), we may write
\[
U = \frac{(2e)^2}{2C} = \frac{2e^2}{C}.
\] (3.2)

We also know that \( Q_j = CV_j \), where \( V_j \) is the voltage of grain \( j \) relative to ground; so
\[
E_{Cj} = \frac{1}{2} CV_j^2 = \frac{Ch^2}{2(2e)^2} \dot{\theta}_j^2,
\] (3.3)
where we have used the Josephson relation, \( V_j = (\hbar/2e) \dot{\theta}_j \). Finally, we can express \( C \) in terms of \( U \) using Eq. (3.2), with the result
\[
E_{Cj} = \frac{\hbar^2}{4U} \dot{\theta}_j^2.
\] (3.4)

Combining all these relations, we obtain
\[
\mathcal{H} = \frac{\hbar^2}{4U} \sum_j \dot{\theta}_j^2 - J \sum_{(ij)} \cos(\theta_i - \theta_j - A_{ij}).
\] (3.5)

We wish to choose the phase factor \( A_{ij} \) in such a way as to include disorder in our model. Since we have taken the grains to lie on a lattice, we need to choose the \( A_{ij} \)'s in a way which incorporates randomness. Thus, we make the simplifying assumption that the phase factor \( A_{ij} \) of each bond in the plane is an independent Gaussian random variable. Since the sum of the phase factors around the four bonds of a plaquette is \( 2\pi/\Phi_0 \) times the flux through that plaquette, this choice will cause the flux through the plaquette also to be a random variable. However, the fluxes through nearest-neighbor plaquettes will be correlated.

Specifically, we assume a Gaussian distribution of each \( A_{ij} \) with a mean of zero and a standard deviation of \( \Delta A_{ij} \). Thus
\[
P(A_{ij}) = \frac{1}{\sqrt{2\pi}(\Delta A_{ij})} \exp \left[-\frac{A_{ij}^2}{2(\Delta A_{ij})^2}\right].
\] (3.6)
We also assume that the values of $A_{ij}$ on different bonds are uncorrelated. Since we are assuming quenched randomness, any given realization of the model will have a particular choice of $A_{ij}$'s chosen from this random distribution.

With this choice of random variables, the model interpolates smoothly between a system in zero magnetic field and a disordered system in a very strong magnetic field. In the first case, $\Delta A_{ij} = 0$ and all the magnetic phase factors $A_{ij} = 0$, corresponding to zero flux through each of the plaquettes, i. e., zero magnetic field. In the second case, $\Delta A_{ij} = \infty$, corresponding to a completely random $A_{ij}$. Since according to the form of the Hamiltonian, $A_{ij}$ and $A_{ij} + 2\pi$ are equivalent, the case $\Delta A_{ij} = \infty$ can be treated by assuming $A_{ij}$ to be randomly distributed between 0 and $2\pi$. We believe that $\Delta A_{ij} = \infty$ should have similar behavior to a random distribution of grains in 2D with a uniform but large magnetic field; in this case, the variations in flux through any given plaquette would be large compared to the average flux. Thus, a model with varying $\Delta A_{ij}$ smoothly between 0 and some very large value should correspond, at least qualitatively, to a disordered system placed in a steadily increasing transverse magnetic field. A small $\Delta A_{ij}$ would represent a disordered 2D system in which the random variations in flux through a plaquette are much less than a single flux quantum, i. e., presumably, a small applied magnetic field, whereas a large $\Delta A_{ij}$ would correspond to the opposite regime.

A somewhat similar way of including random flux has previously been used by Huse and Seung [80] as a model for a 3D “gauge glass.” These workers considered only $\Delta A_{ij} = \infty$, and studied a 3D classical model ($U = 0$).
3.2.2 Path Integral Formulation

We can now use the model Hamiltonian (3.5) to obtain the action in the form of an integral over imaginary time. The action $S$ may be written

$$\frac{S}{\hbar} = \frac{1}{\hbar} \int \mathcal{L} d\tau,$$

(3.7)

where $\mathcal{L}$ is the Lagrangian, given by

$$\mathcal{L} = \frac{\hbar^2}{4U} \sum_j \left( \frac{\partial \theta_j}{\partial \tau} \right)^2 - J \sum_{\langle ij \rangle} \cos[\theta_i(\tau) - \theta_j(\tau) - A_{ij}(\tau)].$$

(3.8)

The partition function is now given by a path integral of $\exp(-S/\hbar)$ over all possible paths described by the variables $\theta_i(\tau)$ in imaginary time $\tau$, integrated from $\tau = 0$ to $\tau = \beta \hbar$, where $\beta = 1/(k_B T)$ and $T$ is the temperature. This path integral can be reduced to the partition function of an anisotropic classical $XY$ model in three dimensions. Here, by “anisotropic” we mean that the coupling constants $K$ and $K_\tau$ in the $xy$ plane and $\tau$ direction are different. To make the mapping, we first write

$$\left( \frac{\partial \theta_i}{\partial \tau} \right)^2 \sim \left( \frac{\Delta \theta_i}{\Delta \tau} \right)^2 \sim \frac{2 - 2 \cos \Delta \theta_i}{(\Delta \tau)^2},$$

(3.9)

where $\Delta \tau$ is the width of the time slice, $\Delta \theta_i = \theta_i(\tau + \Delta \tau) - \theta_i(\tau)$, and we have used the expansion of $\cos \Delta \theta$ to second order in the small quantity $\Delta \theta$. This expansion is accurate when $\Delta \tau$ is sufficiently small.

Neglecting the constant term in this expansion, we finally obtain

$$\frac{S}{\hbar} = -K_\tau \sum\cos[\theta_i(\tau) - \theta_i(\tau + \Delta \tau)] - K \sum\cos[\theta_i(\tau) - \theta_j(\tau) - A_{ij}(\tau)].$$

(3.10)

Here the sums run over all bonds in the $\tau$ direction and in the $xy$ plane, respectively.

In order to obtain the values of the coupling constants $K$ and $K_\tau$, we assume that we have broken up the time integral into $M$ time slices, each of width $\beta \hbar / M$. Then
the coupling constant in the $xy$ direction is just

$$K = \frac{\beta J}{M}. \quad (3.11)$$

The coupling constant in the $\tau$ direction is given by

$$K_\tau = \frac{\hbar^2}{4U} \frac{1}{(\Delta \tau)^2} = \frac{1}{2U} \frac{\hbar}{\Delta \tau} = \frac{M}{2\beta U}, \quad (3.12)$$

we have used $\Delta \tau = \beta \hbar / M$ and included the extra factor of 2 in the numerator because $\cos \Delta \theta \sim 1 - (\Delta \theta)^2 / 2$.

Given $K$ and $K_\tau$, the partition function is obtained using the above anisotropic 3D $XY$ classical Hamiltonian with coupling constants $K$ and $K_\tau$. Any desired equilibrium quantity can, in principle, be computed by averaging over all configurations using standard classical Monte Carlo techniques. Within any given realization of the disorder, the $A_{ij}$’s are chosen at random from the Gaussian distribution within the $xy$ plane, as described above, but the $A_{ij}$’s for a given bond in the $xy$ plane are independent of $\tau$, i.e., they are the same for all time slices. In principle, for any given $\beta$, this should be done taking the limit as $M \to \infty$. In practice, of course, the size of the sample is limited by considerations of computer time.

### 3.2.3 Evaluation of Specific Properties Using Path Integral Formulation

The time-slice formulation of the partition function allows various properties to be evaluated using standard classical Monte Carlo techniques. We now review how this may be done for the helicity modulus (or superfluid density) and the electrical conductivity. Similar formulations have been given in Refs. [29, 30] for different but related models.
Helicity Modulus

For a frustrated classical XY system in \(d\) dimensions, the helicity modulus tensor \(\gamma_{\alpha\beta}\) is a \(d \times d\) matrix which is a measure of the phase stiffness. It is defined as the second derivative of the free energy with respect to an infinitesimal phase twist as in Eq. (2.6). Its diagonal elements are also given in Eq. (2.8).

If that expression is applied to the time-slice representation of the quantum-mechanical Hamiltonian, the coupling constants \(J_{ij}\) will be different in the \(xy\) plane and in the \(\tau\) direction. For the time-slice calculation, we have to be careful in order to obtain a result which is well-behaved in the limit \(M \rightarrow \infty\), where \(M\) is the number of time slices. The correct expression in this case is

\[
\gamma_{xx} = \frac{1}{N_x N_y} \left\langle \frac{J}{M} \sum_{(ij)\parallel\hat{x}} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle 
- \frac{1}{N_x N_y k_B T} \left\langle \left( \sum_{(ij)\parallel\hat{x}} \frac{J}{M} \sin(\theta_i - \theta_j - A_{ij}) \right)^2 \right\rangle 
+ \frac{1}{N_x N_y k_B T} \left\langle \sum_{(ij)\parallel\hat{x}} \frac{J}{M} \sin(\theta_i - \theta_j - A_{ij}) \right\rangle^2. 
\]

(3.13)

Here we are assuming that there are \(N_x N_y\) superconducting grains in our 2D lattice and \(M\) time slices. The sums run over all distinct bonds in the \(\hat{x}\) direction; there are \(N_x N_y M\) of these bonds (\(N_x N_y\) per time slice). A similar expression holds for \(\gamma_{yy}\). The in-plane coupling constant is taken to be \(J/M\) because there are \(M\) time slices.

From the above form, we can see why the expression behaves correctly in the limit \(M \rightarrow \infty\). Each of the two sums contains \(N_x N_y M\) terms in it, but the ensemble average consists of \(M\) identical terms, one for each layer. Therefore, the first sum in

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(3.13), for example, should take the form

$$\frac{J}{M} \left\langle \sum_{(ij)} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle \rightarrow J \left\langle \sum'_{(ij)} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle,$$

(3.14)

where the sum on the right hand side runs only over the phases in a single layer. The right-hand side is evidently independent of $M$ in the limit $M \to \infty$. A similar argument can be used to show that the second part of the expression (3.13) for $\gamma_{xx}$ also approaches a well-behaved limit as $M \to \infty$. Our numerical results confirm this behavior.

As an illustration, we write down an expression for $\gamma_{xx}$ in the limit $T \to 0$ in the unfrustrated case ($\Delta A_{ij} = 0$). First, we multiply expression (3.13) by $\beta/M$ to obtain

$$\frac{\beta \gamma_{xx}}{M} = \frac{1}{N_x N_y M} \left\langle K \sum_{(ij)} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle$$

$$- \frac{1}{N_x N_y M} \left\langle \left( \sum_{(ij)} K \sin(\theta_i - \theta_j - A_{ij}) \right)^2 \right\rangle$$

$$+ \frac{1}{N_x N_y M} \left\langle \left( \sum_{(ij)} K \sin(\theta_i - \theta_j - A_{ij}) \right)^2 \right\rangle,$$

(3.15)

where $K = \beta J/M$. The corresponding coupling constant in the $\tau$ direction is $K_\tau = M/(2\beta U)$.

Since we are interested in the limit $\beta \gg 1$, we will choose $M$ so that $K = K_\tau$. This condition is equivalent to

$$M = \beta \sqrt{2JU},$$

(3.16)
or equivalently $\beta/M = 1/\sqrt{2JU}$. Hence, we get

$$\frac{\gamma_{xx}}{\sqrt{2JU}} = \frac{1}{N_x N_y M} \left\langle K \sum_{\langle ij \rangle \parallel \hat{x}} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle$$

$$- \frac{1}{N_x N_y M} \left\langle \left( \sum_{\langle ij \rangle \parallel \hat{x}} K \sin(\theta_i - \theta_j - A_{ij}) \right)^2 \right\rangle$$

$$+ \frac{1}{N_x N_y M} \left\langle \left( \sum_{\langle ij \rangle \parallel \hat{x}} K \sin(\theta_i - \theta_j - A_{ij}) \right)^2 \right\rangle, \quad (3.17)$$

where $K = \beta J/M = \sqrt{J/(2U)}$. Since $K = K_{\gamma}$, the right-hand side of this equation represents a dimensionless helicity modulus $\tilde{\gamma}$ for a classical unfrustrated isotropic 3D $XY$ model on a simple cubic lattice, which is a function of a single dimensionless coupling constant $K$. $K$ represents the ratio of the coupling strength $J_{\text{eff}}$ of that isotropic 3D $XY$ model to $k_B T$.

Now it is known from previous Monte Carlo studies [111, 65, 52, 154, 149] that the unfrustrated 3D $XY$ model on a simple cubic lattice has an ordered phase if $K > K_c \approx 1/2.21 \approx 0.452$. Therefore, $\tilde{\gamma}(K)$ vanishes if $K < K_c$ and is positive for $K > K_c$. Translating this result to the 2D quantum $XY$ model on a square lattice, we see that there is a superconductor–insulator transition at $J/(2U) = 0.452^2 = 0.204$.

For reference we give the connection between our formulation of the helicity modulus and the calculation of Cha et al. [29]. Rather than the helicity modulus, these workers calculate the quantity $\rho(0)$, which is related to the superfluid density $\rho_s$ by

$$\rho(0) = \frac{\rho_s}{k_B T}, \quad (3.18)$$

and to the components of the helicity modulus tensor by $\rho(0) = K \gamma \equiv K(\gamma_{xx} + \gamma_{yy})/2$, where $\gamma_{xx} = \gamma_{yy}$ for the present model, which is isotropic in the $xy$ plane. In our
notation, \( \rho(0) \) is given by

\[
\rho(0) = \frac{JK}{2N_xN_yM}\left[ \left< \sum_{(ij)\parallel \hat{x}} \cos(\theta_i - \theta_j - A_{ij}) \right> + \left< \sum_{(ij)\parallel \hat{y}} \cos(\theta_i - \theta_j - A_{ij}) \right> \right]
- \frac{JK^2}{2N_xN_yM}\left[ \left< \left[ \sum_{(ij)\parallel \hat{x}} \sin(\theta_i - \theta_j - A_{ij}) \right]^2 \right> \right]
+ \left< \left[ \sum_{(ij)\parallel \hat{y}} \sin(\theta_i - \theta_j - A_{ij}) \right]^2 \right> \right]
+ \left< \left[ \sum_{(ij)\parallel \hat{y}} \sin(\theta_i - \theta_j - A_{ij}) \right]^2 \right> .
\] (3.19)

**Specific Heat**

For the specific heat \( C_V \), we used the fluctuation-dissipation theorem given in Eq. (2.4) replacing \( \mathcal{H} \) with \( \mathcal{H}' \), which is the Hamiltonian in Eq. (3.5).

**Conductivity**

The conductivity in the low-frequency limit can also be obtained from the time-slice Monte Carlo approach as [29]

\[
\sigma(0) = 2\pi\sigma_Q \lim_{k \to 0} \frac{\rho(k)}{k} ,
\] (3.20)
where $\rho(k)$ is given by

$$
\rho(k) = \frac{JK}{2N_xN_yM} \left[ \left\langle \sum_{\langle ij \rangle \parallel \hat{x}} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle + \left\langle \sum_{\langle ij \rangle \parallel \hat{y}} \cos(\theta_i - \theta_j - A_{ij}) \right\rangle \right]
$$

$$
- \frac{JK^2}{2N_xN_yM} \left\langle \sum_{\langle ij \rangle \parallel \hat{x}} \sin(\theta_i - \theta_j - A_{ij})e^{-ik\cdot\hat{x}} \sum_{\langle ij \rangle \parallel \hat{x}} \sin(\theta_i - \theta_j - A_{ij})e^{ik\cdot\hat{x}} \right\rangle
$$

$$
- \frac{JK^2}{2N_xN_yM} \left\langle \sum_{\langle ij \rangle \parallel \hat{y}} \sin(\theta_i - \theta_j - A_{ij})e^{-ik\cdot\hat{y}} \sum_{\langle ij \rangle \parallel \hat{y}} \sin(\theta_i - \theta_j - A_{ij})e^{ik\cdot\hat{y}} \right\rangle
$$

$$
+ \frac{JK^2}{2N_xN_yM} \left\langle \sum_{\langle ij \rangle \parallel \hat{y},\hat{x}} \sin(\theta_i - \theta_j - A_{ij})e^{-ik\cdot\hat{y}} \right\rangle
$$

$$
\times \left\langle \sum_{\langle ij \rangle \parallel \hat{x}} \sin(\theta_i - \theta_j - A_{ij})e^{ik\cdot\hat{x}} \right\rangle
$$

$$
+ \frac{JK^2}{2N_xN_yM} \left\langle \sum_{\langle ij \rangle \parallel \hat{x}} \sin(\theta_i - \theta_j - A_{ij})e^{-ik\cdot\hat{x}} \right\rangle
$$

$$
\times \left\langle \sum_{\langle ij \rangle \parallel \hat{y}} \sin(\theta_i - \theta_j - A_{ij})e^{ik\cdot\hat{y}} \right\rangle. \quad (3.21)
$$

In the limit of very small $k$, we expect that $\rho(k)$ will remain finite in the superconducting phase and vanish in the insulating phase. Thus $\sigma(0)$ will become infinite in the superconducting phase but vanish in the insulating phase. Precisely at the critical value $K_c$, $\sigma(0)$ will become finite with a universal value, as already obtained by other workers for related model.

### 3.3 Quantum Monte Carlo Results

#### 3.3.1 Numerical Procedure

In our quantum Monte Carlo calculations, we use the standard Metropolis algorithm with periodic boundary conditions in both the spatial directions and the imaginary time direction. We usually start with a random configuration of phases at $K = 0.4$, then increase $K$ up to $K = 0.7$ in steps of 0.005. This procedure corresponds
to lowering the temperature $T$ since $K \propto 1/T$. At each $K$, we take 40000 MC steps per site through the entire lattice to equilibrate the system, after which we take an additional 50000 MC steps to calculate the thermodynamic variables of interest. For a lattice size of $6^3$, we use ten times as many MC steps as these for both equilibration and averaging, and for a lattice size of $8^3$, we use twice as many steps.

For the phases $\theta_i$ of the order parameter on each site, we use the 360-state clock model instead of a continuous angles between 0 and $2\pi$ as we did in Sec. 2.3. However, we select $A_{ij}$ from a continuous distribution in all our calculations.

For the partially random and completely random $A_{ij}$, we averaged over 100 different realizations of $A_{ij}$ to calculate the helicity modulus $\gamma$ and the specific heat $C_V$. These calculations were so time-consuming that we could go just up to $12 \times 12 \times 12$ lattice size compared to the $20 \times 20 \times 20$ lattice size when $A_{ij} = 0$. For this reason, we chose to carry out simulations only over four different $\Delta A_{ij}$ for the partially random case: $\Delta A_{ij} = 1/2, 1/\sqrt{2}, (1 + 1/\sqrt{2})/2 \approx 0.854$, and 1. For each realization, we fixed the seed of the random number generator, so that we could reproduce the same array of $A_{ij}$ for different $\Delta A_{ij}$ for each lattice size.

### 3.3.2 Finite-Size Scaling for $\gamma(0)$

In general, if there is a continuous phase transition as a function of some parameter, such as the coupling constant $K$, the critical behavior near the transition can be analyzed by carrying out a finite-size scaling analysis of various calculated quantities. For example, the frequency-independent helicity modulus $\gamma(0)$ is expected to satisfy [183]

$$
\gamma(0) = \frac{1}{L^{d+z-2}} \tilde{\gamma} \left( L^{1/\nu} \delta \frac{L_\tau}{L^z} \right),
$$

(3.22)
where \( d \) is the spatial dimensionality, \( z \) is the dynamic exponent, \( \tilde{\gamma} \) is a scaling function, \( \nu \) is the critical exponent of the correlation length \( \xi \), \( \delta = (K - K_c)/K_c \), \( K_c \) is the critical value of the coupling constant, and \( L_r \) is the lattice size in the imaginary time direction. Generally, \( L_r = cL^z \), where \( c \) is the aspect ratio. In the present chapter we assume \( N_x = N_y = M \). Therefore Eq. (3.22) simplifies to

\[
L^z\gamma(0) = \tilde{\gamma}(L^{1/\nu}\delta)
\]

(3.23)

when \( d = 2 \).

### 3.3.3 Zero Magnetic Field

We first calculated the helicity modulus \( \gamma \) and the specific heat \( C_V \) as functions of charging energy \( U \) for three different lattice sizes, \( 16 \times 16 \times 16, 18 \times 18 \times 18, \) and \( 20 \times 20 \times 20 \) when there is no magnetic field \( (A_{ij} = 0) \) as shown in Fig. 3.1. Here we assumed \( \beta = 10.0 \). As the lattice size increases, \( U_c \) decreases because \( K \propto 1/M \). The critical charging energy \( U_c \) from \( \gamma \) well matches the peak of \( C_V \) curves.

Fig. 3.2 is the same as Fig. 3.1, except that it is plotted as functions of \( T' = 1/\beta \) with \( U = 4.0 \). Here we can see the clear effect of quantum fluctuations because the helicity modulus (or superfluid density) doesn’t drop sharply at \( T_c \), but drops smoothly to zero. Again, the critical temperature \( T'_c \) from \( \gamma \) well matches the peak of \( C_V \) curves.

Then we calculated \( K_c \) in zero magnetic field. In this case, we get \( K_c = 0.4543 \pm 0.0011 \) using a finite-size scaling analysis of the helicity modulus \( \gamma \) as shown in Fig. 3.3. Also the PRG method in Sec. 2.2 is used in Fig. 3.4, which gives \( K_c = 0.4542 \). These values are very close to \( K_c = 0.4539 \pm 0.0013 \) by the series expansion as in Ref. [51], which is also used in Ref. [29]. These results confirm the validity of our
Figure 3.1: (a) Helicity modulus $\gamma$ and (b) specific heat $C_V$, plotted as functions of charging energy $U$ for three different lattice sizes, $16 \times 16 \times 16$, $18 \times 18 \times 18$, and $20 \times 20 \times 20$ when $\beta = 10$ and $A_{ij} = 0$. The error bars in (a), as obtained from the jackknife method, are smaller than the symbol sizes. The lines in (b) are cubic spline fits to the Monte Carlo data.

3.3.4 Finite $\Delta A_{ij}$

Figs. 3.6(a), 3.6(b), and 3.7 show the helicity modulus $\gamma$, the specific heat $C_V$, and the finite-size scaling behavior of $\gamma$ as a function of coupling constant $K$ for several lattice sizes when $\Delta A_{ij} = 1/2$. When $K > 0.55$, $\gamma$ and $C_V$ are mostly lattice size independent. The error bars from the jackknife method [127] are shown in Fig. 3.6(a), but they are smaller than the symbol sizes. The lines are cubic spline fits to the data in Fig. 3.6(b). The crossing point in Fig. 3.7 yields the critical coupling constant $K_c = 0.490 \pm 0.001$, which is very close to the peak of $C_V$ in Fig. 3.6(b).

numerical codes. Using our value of $K_c$, we can obtain the universal conductivity $\sigma^*/\sigma_Q = 0.282 \pm 0.005$ as shown in Fig. 3.5. This result is also very close to the value $\sigma^*/\sigma_Q = 0.285 \pm 0.02$ obtained in Ref. [29].
Figure 3.2: Same as Fig. 3.1, except that they are plotted as functions of $T' = 1/\beta$ when $U = 4.0$.

Figure 3.3: Plot of $\gamma L_\tau$ as a function of $K$ for various $L \times L \times L_\tau$ lattices of size, for $A_{ij} = 0$. The phase transition occurs where the curves of different $L$ cross. The crossing point yields $K_c = 0.4543 \pm 0.0011$. 
Figure 3.4: PRG method to extract the critical coupling constant $K_c$ from the data used in Fig. 3.3. All three curves show $\gamma(L, K)/\gamma(L', K)$ for the same ratio of $L/L' = 2$. The abscissa of the intersection of the three curves yields $K_c = 0.4542$.

The corresponding results for $\Delta A_{ij} = 1/\sqrt{2}$ are shown in Figs. 3.8(a), 3.8(b), and 3.9. Unlike Fig. 3.6(a) $\gamma$ shows some lattice size dependence when $K > K_c$ in Fig. 3.8(a). The crossing point in Fig. 3.9 yields $K_c = 0.532 \pm 0.001$. This $K_c$ is also very close to the peak in $C_V$ as in Fig. 3.8(b).

The results for $\gamma$, $C_V$, and $\gamma L_\tau$ when $\Delta A_{ij} = 0.854$ are shown in Figs. 3.10(a), 3.10(b), and 3.11. In this case, the lattice size-dependence of $\gamma$ when $K > K_c$ in Fig. 3.10(a) becomes more conspicuous than that of Fig. 3.8(a). The crossing point for the different sizes in Fig. 3.11, though not quite so clearly defined as in the previous examples, yields $K_c = 0.585 \pm 0.004$. This $K_c$ is slightly larger than the value of $K$ at the maximum of the broad peak in $C_V$, as in Fig. 3.10(b). There are some fluctuations of $\gamma L_\tau$ around $K = 0.70$ for the lattice size of $12^3$ in Fig. 3.11.
Figure 3.5: The conductivity $\sigma(n, L/\alpha n)$ divided by $\sigma_Q$ as a function of the variable $\alpha/n - n/L_\tau$ for several lattice sizes $L$ when $A_{ij} = 0$ and $K = K_c = 0.4542$. The optimal $\alpha$ used here is 0.74. The universal conductivity $\sigma^*/\sigma_Q$ is given by that value of $\sigma(n, L/\alpha n)$ obtained when $\alpha/n - n/L_\tau = 0$, as indicated by the vertical dotted line. The universal conductivity thus obtained is $\sigma^*/\sigma_Q = 0.282 \pm 0.005$.

As a final calculation for partially random $A_{ij}$, we use $\Delta A_{ij} = 1.0$. The corresponding three thermodynamic variables $\gamma$, $C_V$, and $\gamma L_\tau$ are shown in Figs. 3.12(a), 3.12(b), and 3.13, respectively. The lattice size-dependence of $\gamma$ in Fig. 3.12(a) becomes far more conspicuous than the previous two examples. The peak of $C_V$ is very broad, as shown in Fig. 3.12(b). In addition, there is no clear crossing point of $\gamma L_\tau$ for different sizes $L$ in Fig. 3.13. We interpret this result to mean that the helicity modulus $\gamma$ no longer plays the role of an order parameter, and hence that the transition is no longer a superconductor-to-insulator transition of the same character as for smaller values of $\Delta A_{ij}$. Furthermore, there are strong fluctuations of $\gamma L_\tau$ as a function of $L$ when $K \geq 0.64$ for most lattice sizes, as can be seen in Fig. 3.13. We believe that, for this value of $\Delta A_{ij}$, this is a transition from a Bose glass to a
Figure 3.6: (a) Helicity modulus $\gamma$ and (b) specific heat $C_V$, plotted as functions of coupling constant $K$ for several lattice sizes when $\Delta A_{ij} = 1/2$. The error bars in (a), as obtained from the jackknife method, are smaller than the symbol sizes. The lines in (b) are cubic spline fits to the Monte Carlo data.

Figure 3.7: Finite-size scaling behavior of the data in Fig. 3.6(a). The crossing point yields $K_c = 0.490 \pm 0.001$. 
Figure 3.8: Same as Fig. 3.6, except that $\Delta A_{ij} = 1/\sqrt{2}$.

Figure 3.9: Same as Fig. 3.7, except that the data are from Fig. 3.8(a). The crossing point yields $K_c = 0.532 \pm 0.001$, as indicated by the vertical dashed line.

Mott insulator and conclude that the transition changes from the superconductor-to-insulator to the Bose-glass-to-insulator in the region $0.854 < \Delta A_{ij} < 1.0$. This interpretation is discussed further below.
Figure 3.10: Same as Fig. 3.6, except that $\Delta A_{ij} = 0.854$.

Figure 3.11: Same as Fig. 3.7, except that the data are from Fig. 3.10(a). The crossing point yields $K_c = 0.585 \pm 0.004$.

Finally, we have considered the case of a fully random $A_{ij}$, $\Delta A_{ij} = \infty$. The helicity modulus $\gamma$ and the specific heat $C_V$ for this case are shown in Figs. 3.14(a)
Figure 3.12: Same as Fig. 3.6, except that $\Delta A_{ij} = 1.0$.

Figure 3.13: Same as Fig. 3.7, except that the data are from Fig. 3.12(a). In this case, the plots of $L_\tau \gamma(K)$ for different $L$ do not cross, suggesting that the helicity modulus $\gamma$ is no longer a suitable order parameter at $\Delta A_{ij} = 1.0$. 
and 3.15(a), respectively. The magnitude of $\gamma$ becomes much smaller than those of previous cases, so that the error bars are easily visible on the scale of the plot. The helicity modulus even seems to have negative values for certain values of $K$, depending on the lattice size. Such negative values and fluctuations of the helicity modulus in a disordered superconductor were already reported in other work [163], in the context of a different model. As in Figs. 3.12(a) and 3.13, $\gamma$ is strongly lattice-size dependent and there exists no value of $K$ at which the curves of $L_x\gamma(K)$ for different $L$ all cross (we do not show a plot exhibiting this lack of crossing). All these results suggest that we need a different order parameter to describe the phase transition when $\Delta A_{ij} = \infty$. Besides these results, we find that the peak in $C_V(K)$ is even broader than that in Fig. 3.12(b). Moreover, the peak of $C_V$ shifts towards a larger value of $K$ as $\Delta A_{ij}$ increases.

As a comparison to the fully random $A_{ij}$ case, we have also considered a model of "positionally disordered sites" in a strong uniform transverse magnetic field $B = B\hat{z}$, similar to a model considered in Ref. [155]. The position coordinates $(x_i, y_i)$ of each site are assumed uniformly and independently distributed between $-\Delta$ and $\Delta$ with respect to the position $(x_{i0}, y_{i0})$ the site would have in the ordered lattice, i.e.,

\[
|x_i - x_{i0}| \leq \Delta, \\
|y_i - y_{i0}| \leq \Delta. 
\]  

(3.24)

In our calculations, we have chosen $\Delta = a/4$, where $a$ is the lattice constant of the unperturbed lattice. Thus $A_{ij}$ has the form

\[
A_{ij} = \frac{2\pi}{\Phi_0} B \frac{x_i + x_j}{2} (y_j - y_i) 
\]  

(3.25)
for nearest-neighbor sites $i$ and $j$. In order to consider a strong field, we choose $f = Ba^2/\Phi_0 = 20$. The results for this system of positionally disordered sites are shown in Fig. 3.14(b) and 3.15(b). They are qualitatively similar to those with $\Delta A_{ij} = \infty$, even quantitatively for $C_V$. We conclude that the model of positionally disordered sites is nearly equivalent to that with random $A_{ij}$, at least for $\Delta A_{ij} = \infty$.

At large values of $\Delta A_{ij}$, our results suggest that the transition occurs between a Mott insulator and a Bose glass rather than a conventional superconductor. Since a new order parameter is demanded to study this transition, we use the “renormalized coupling constant $g$” as in Ref. [80]. Using the same $A_{ij}$ for each realization, two replicas of phase $\theta_j$ are simulated with different initial conditions and updated using
Figure 3.15: Same as Fig. 3.14 except both for the specific heat $C_V$. The lines are cubic spline fits to the data.

different random numbers. Their overlap is calculated from the quantity

$$q = \sum_j \exp[i(\theta_j^{(1)} - \theta_j^{(2)})],$$  \hspace{1cm} (3.26)

where $\theta_j^{(1)}$ and $\theta_j^{(2)}$ are the phases at site $j$ in the two replicas. Given $q$, the renormalized coupling constant $g$ is defined as

$$g = 2 - \frac{[\langle |q|^4 \rangle]}{[\langle |q|^2 \rangle]^2},$$  \hspace{1cm} (3.27)

where $\langle \cdots \rangle$ denotes the thermal average while $[\cdots]$ denotes an average over many realizations of $A_{ij}$. Figure 3.16 shows this $g$ as a function of coupling constant $K$ for several lattice sizes when $\Delta A_{ij} = \infty$. From the crossing point for different sizes, we obtain $K_c = 0.630 \pm 0.002$. Unlike the results in Ref. [80], $g$ still has a size dependence when $K > K_c$. We speculate that this size dependence results from the additional charging energy term in our model, as given in Eq. (3.5).
Figure 3.16: The renormalized coupling constant $g$ [Eq. (3.27)] as a function of coupling constant $K$ for several lattice sizes when $\Delta A_{ij} = \infty$. The crossing point yields $K_c = 0.630 \pm 0.002$. The lines are cubic spline fits to the data.

With all the $K_c$'s we have collected so far, we can plot $1/K_c$ as a function of $\Delta A_{ij}$. This is shown in Fig. 3.17. Since $K_c = \sqrt{[J/(2U)]_c}$ and since $1/K_c$ decreases as $\Delta A_{ij}$ increases, these results mean that $[J/(2U)]_c$ increases with increasing $\Delta A_{ij}$. Therefore, there exist certain values of the ratio $J/U$ such that the system is superconducting for small $\Delta A_{ij}$ but insulating for large $\Delta A_{ij}$. If we interpret $\Delta A_{ij}$ as representing a quantity similar to a uniform magnetic field in a disordered system, then, for certain values of $J/U$, the system undergoes a superconductor-to-insulator transition as a function of magnetic field. Such transitions have been observed in several experiments, as discussed earlier. Since $J/U$ is presumably a fixed quantity for a given material, this means that there should be some materials which undergo such field-dependent S-I transitions.
Figure 3.17: Inverse critical coupling constant $K_c$ as a function of $\Delta A_{ij}$. The solid line and the dashed line are cubic spline fits to the data. ‘$S$’ denotes a superconducting phase, ‘$I$’ denotes an insulating phase, and ‘$BG$’ denotes a Bose glass phase.

In Fig. 3.17, we have shown the possible phase boundaries among a superconductor, a Mott insulator, and a Bose glass. The phase boundary between the superconductor and the Bose glass is not clear because the transition occurs in the region $0.854 < \Delta A_{ij} < 1.0$. Above this region, as mentioned above, the ordered system is no longer a superconductor with a superfluid density that obeys conventional scaling. Instead, it appears to be a glassy phase, which we interpret as a Bose glass, since it is disordered in the $xy$ plane but ordered in the imaginary time direction. We expect that the ordered phase will have this Bose glass character for sufficiently high fields. Thus, we might have the exotic possibility that, for certain values of $J/U$, the material might undergo two transitions as a function of magnetic field, from superconductor to Bose glass to insulator.
### 3.3.5 Conductivity

In order to obtain the value of the conductivity at the transition, we need to generalize the scaling formulas to frequency-dependent $\gamma$. When there is such a frequency dependence, Eq. (3.23) is generalized to [29]

$$L^z\gamma(k) = \tilde{\gamma}(L^{1/\nu}kL_\tau), \quad (3.28)$$

where $k = 2\pi n/L_\tau$ and $n$ is an integer. Precisely at $K = K_c$, $\tilde{\gamma}$ will be a function of only $kL_\tau$ since $K - K_c = 0$. Thus we can introduce another scaling function $P$, in terms of which Eq. (3.28) can be simplified to

$$L^z\gamma(k) = P(kL_\tau). \quad (3.29)$$

From Eq. (3.20), the conductivity is obtained by taking the limit $k \to 0$ after first taking the limit $L_\tau \to \infty$ with a small $k$ [29], so that $P(kL_\tau) \simeq kL_\tau$ in the limit $L_\tau \to \infty$. Using the scaling function $P$, Eq. (3.20) can be rewritten as [29]

$$\frac{\sigma^*}{\sigma_Q} = 2\pi \lim_{kL_\tau \to \infty} \frac{P(kL_\tau)}{kL_\tau}. \quad (3.30)$$

Since this quantity is to be calculated for $k \to 0$ after $L_\tau \to \infty$, the ratio $\sigma^*/\sigma_Q$ will be finite only if the scaling function $P(x) \propto x$ in this regime. Since $k = 2\pi n/L_\tau$, the scaling form (3.30) can be written again as [29]

$$\frac{\sigma(n)}{\sigma_Q} = \frac{P(2\pi n)}{n}. \quad (3.31)$$

This scaling form is expected to be valid only in the regime $1 \ll n \ll L_\tau$. Since it is difficult to carry out calculations for $L_\tau$ large enough that these inequalities are satisfied, especially for a disordered system, it is necessary to incorporate corrections to scaling and express $\sigma$ in terms of $n$ and $L_\tau/n$ separately. Since the corrections to
scaling vanish in the limit \( L_\tau /n \to \infty \), we expand \( \sigma(n) \) as a function of \( n \) and \( L_\tau /n \)
using the same form assumed in Ref. [29], namely
\[
\frac{\sigma(n, L_\tau /n)}{\sigma_Q} = \frac{\sigma^*}{\sigma_Q} + d \left( \frac{\alpha}{n} - \frac{n}{L_\tau} \right) + \cdots ,
\] (3.32)
where \( d \) and \( \alpha \) are fitting constants. The universal conductivity \( \sigma^* \) is found by plotting \( \sigma(n, L_\tau /n) \) as a function of the scaling variable \( (\alpha/n - n/L_\tau) \) for several lattice sizes \( L \) and finding the optimal value of \( \alpha \) which produces the best data collapse onto a single curve. The universal conductivity for this value of \( \Delta A_{ij} \) is the value of \( \sigma^* \) at which \( \alpha/n - n/L_\tau = 0 \).

Using this method, we find the following universal conductivities for different values of \( \Delta A_{ij} \): \( \sigma^*/\sigma_Q = 0.324 \pm 0.003 \) when \( \Delta A_{ij} = 1/2 \), \( \sigma^*/\sigma_Q = 0.494 \pm 0.011 \) when \( \Delta A_{ij} = 1/\sqrt{2} \), \( \sigma^*/\sigma_Q = 0.973 \pm 0.068 \) when \( \Delta A_{ij} = 0.854 \), and \( \sigma^*/\sigma_Q = 0.896 \pm 0.085 \) when \( \Delta A_{ij} = \infty \). At each of these values of \( \Delta A_{ij} \), we apply the method just described to calculate the universal conductivity at the corresponding \( K_c \) values obtained earlier. The results are shown in Figs. 3.18, 3.19, 3.20, and 3.21, respectively. The optimal values of \( \alpha \)'s which yield these universal conductivities are \( \alpha = 0.63 \), 0.29, 0.08 and 0.02 for \( \Delta A_{ij} = 1/2 \), \( \Delta A_{ij} = 1/\sqrt{2} \), \( \Delta A_{ij} = 0.854 \), and \( \Delta A_{ij} = \infty \), respectively. The accuracy of the calculated \( \sigma^*/\sigma_Q \) becomes progressively worse as \( \Delta A_{ij} \) increases. In fact, we need to obtain the results for \( \Delta A_{ij} = 0.854 \) and \( \Delta A_{ij} = \infty \) by extrapolation of \( \sigma(n, L_\tau /n)/\sigma_Q \) to the optimal values of \( \alpha \), using Figs. 3.20 and 3.21.

The critical coupling constant \( K_c \) increases monotonically with increasing \( \Delta A_{ij} \) in both the superconductor-to-insulator and Bose-glass-to-insulator transition regimes. By contrast, the universal conductivity \( \sigma^* \) increases monotonically with \( \Delta A_{ij} \) for the superconductor-to-insulator transition, but decreases with increasing \( \Delta A_{ij} \) for the Bose-glass-to-insulator transition. This behavior is not surprising, because, as
Figure 3.18: The conductivity $\sigma(n, L_r/n)$ divided by $\sigma_Q$ as a function of the variable $\alpha/n - n/L_r$ for several lattice sizes $L$ when $\Delta A_{ij} = 1/2$ and $K = K_c = 0.490$. The optimal $\alpha$ used here is 0.63. The universal conductivity $\sigma^*/\sigma_Q$ is given by that value of $\sigma(n, L_r/n)$ obtained when $\alpha/n - n/L_r = 0$, as indicated by the vertical dashed line. The universal conductivity thus obtained is $\sigma^*/\sigma_Q = 0.324 \pm 0.003$.

Previously found in Ref. [30] (using a different model from ours), the value $\sigma^*$ for the Bose-glass-to-insulator transition is lower than that for the superconductor-to-insulator transition. It reported that $\sigma^*/\sigma_Q = 0.52 \pm 0.03$ for a disorder-free model with the frustration $f = 1/2$ and that $\sigma^*/\sigma_Q = 0.49 \pm 0.04$ for a disordered model with $f = 1/2$.

From these universal conductivities, we can plot the universal resistivities as a function of $\Delta A_{ij}$. These are shown in Fig. 3.22. The resistivity decreases with increasing $\Delta A_{ij}$ along the phase boundary between a superconductor and a Mott insulator, while it slightly increases along that between a Bose glass and the Mott insulator. The solid line and the dashed line represent the cubic spline fits to the data, which
Figure 3.19: Same as Fig. 3.18, except that $\Delta A_{ij} = 1/\sqrt{2}$ and $K_c = 0.532$. The optimal $\alpha$ used here is 0.29. The universal conductivity is $\sigma^*/\sigma_Q = 0.494 \pm 0.011$.

Figure 3.20: Same as Fig. 3.18, except that $\Delta A_{ij} = 0.854$ and $K_c = 0.585$. The optimal $\alpha$ used here is 0.08. We therefore have to extrapolate the plot of the conductivity $\sigma(n, L/n)/\sigma_Q$ to reach the point at $\alpha/n - n/L_\tau = 0$. The universal conductivity thus obtained is $\sigma^*/\sigma_Q = 0.973 \pm 0.068$. 
are shown as filled small circles. Again the phase boundary between the superconductor and the Bose glass is not exact because it’s in the region $0.854 < \Delta A_{ij} < 1.0$, above which the system, as already mentioned, appears to undergo a Bose glass to an insulator transition rather than a superconductor-to-insulator transition.

If we interpret increasing $\Delta A_{ij}$ as corresponding roughly to increasing a uniform magnetic field applied to a disordered sample, then our results, which show a decrease in the universal resistivity with increasing $\Delta A_{ij}$, may be qualitatively consistent with the experiments reported in Refs. [118, 120] and with the previous numerical results as in Ref. [134]. However, these experiments are not equivalent to our simplified model, since they consider a disordered Bi film in a uniform magnetic field, rather than a periodic Josephson junction array with a fluctuating field. The experiments study the zero-temperature magnetic-field-tuned superconductor–insulator transition, and
interpret them by carrying out scaling fits of the low-temperature transport properties near the critical field, as in the present chapter.

### 3.4 Discussion

In this chapter, we have calculated the transition between the superconducting and the insulating state for a model disordered 2D superconductor in a magnetic field. We model the superconductor as a 2D Josephson junction array with an intergranular Josephson coupling energy $J$ and a finite capacitive energy described by an on-site charging energy $U$. In order to model the effect of a magnetic field, we assume an ordered array and describe the field by a random magnetic phase factor $A_{ij}$ in the Josephson coupling between grains; $A_{ij}$ is assumed to have a Gaussian distribution.
with zero mean and a root-mean-square width $\Delta A_{ij}$. This way of approach to the 2D S-I transition has never been performed before.

Although our model is certainly artificial, we argue that it does provide a reasonable representation of the effects of a magnetic field applied to a real disordered 2D film. Specifically, in our model, the limit $\Delta A_{ij} = 0$ corresponds to zero applied magnetic field, whereas for very large $\Delta A_{ij}$, the flux through a plaquette of grains is very widely distributed. Thus, in these two limits, the model gives a reasonable representation of a spatially disordered 2D granular superconductor in a uniform magnetic field. In particular, it includes the important ingredient of plaquettes of grains which are randomly frustrated. Our model does provide a way of interpolating smoothly between these limits. In particular, in the intermediate $\Delta A_{ij}$ regime, it includes the effects of partial frustration which may be important in a realistic 2D film.

Our central result is that, for any value of $\Delta A_{ij}$ at temperature $T = 0$, we find a transition from an insulating state to an ordered state as a function of the coupling parameter $K = \sqrt{J/(2U)}$ which describes the ratio of Josephson coupling to charging energy in our model. For small $\Delta A_{ij}$, this is a transition from the insulating to a superconducting state described by a finite superfluid density. For large $\Delta A_{ij}$, the ordered state appears to be a Bose glass rather than a superconducting state. This glass phase appears to have zero superfluid density. Instead it has a nonzero “renormalized coupling constant $g_r$,” which has previously been used to treat glass phases in other models [80].

We find that the critical value $K_c$ of the coupling for a transition into the ordered state is a monotonically increasing value of $\Delta A_{ij}$. Since for any given material, the ratio $K$ is presumably independent of magnetic field, this result means that, for some
values of $K$ (i.e., for certain materials), as $\Delta A_{ij}$ increases, the system will undergo a transition from superconducting to insulating. For larger values of $K$, the system will undergo a superconducting to Bose glass transition, and for some values there may be two transitions, from superconducting to Bose glass to insulating. Finally, for small enough values of $K$, the system will be insulating for all values of $\Delta A_{ij}$. The insulating phase in our model is basically a Mott insulator since Cooper pairs are localized by Coulomb repulsion rather than by disorder.

Besides calculating the critical values of coupling constant $K_c$, we have also computed the universal conductivities $\sigma^*$ as a function of $\Delta A_{ij}$ for these transitions. We find $K_c = 0.490 \pm 0.001$ and $\sigma^*/\sigma_Q = 0.324 \pm 0.003$ when $\Delta A_{ij} = 1/2$; $K_c = 0.532 \pm 0.001$ and $\sigma^*/\sigma_Q = 0.494 \pm 0.011$ when $\Delta A_{ij} = 1/\sqrt{2}$; $K_c = 0.585 \pm 0.004$ and $\sigma^*/\sigma_Q = 0.973 \pm 0.068$ when $\Delta A_{ij} = 0.854$; and $K_c = 0.630 \pm 0.002$ and $\sigma^*/\sigma_Q = 0.896 \pm 0.085$ when $\Delta A_{ij} = \infty$. These values are obtained by a scaling analysis of the numerically calculated helicity modulus $\gamma$ and the renormalized coupling constant $g$. We conclude that the 2D disordered superconducting films undergo a phase transition from a superconductor to a Mott insulator up to $K_c = 0.854$ and a phase transition from a Bose glass to a Mott insulator above $K_c = 0.854$. $K_c$ increases monotonically with $\Delta A_{ij}$, as shown in Fig. 3.17. By contrast, $\sigma^*$ increases monotonically with increasing $\Delta A_{ij}$ for superconductor-to-insulator transition, but appears to decreases monotonically with increasing $\Delta A_{ij}$ for the Bose-glass-to-insulator transition. However, we have very limited data in the latter regime.

These results are consistent with experimental findings as in Refs. [73, 192, 118, 120]. Numerically, there are several ways in which our calculations could be further improved. In some cases, the number of realizations (100) we have used for $A_{ij}$ might...
not be sufficient to provide accurate statistics and might lead to significant numerical uncertainties in the statistics. Our choice for the number of realizations is dictated by a compromise between computing costs and statistical errors. Because of the large amount of computing time involved, we have carried out our calculations only up to a lattice size of $12 \times 12 \times 12$, and have considered only five different nonzero $\Delta A_{ij}$’s. Our results would have had greater accuracy and given a more detailed picture of the phase diagram if we had been able to include a larger number of realizations, larger lattice sizes, and more values $\Delta A_{ij}$. In addition, our “world-line” algorithm [78, 12, 11, 10] could be replaced by other approaches, such as a “worm” algorithm [1, 2] or a stochastic series expansion [152, 75, 194, 160].

Throughout this chapter, we have taken the aspect ratio $c = L_r/L^z$ for our Monte Carlo cell size to be a constant. In general, in most previous work, such as Refs. [162, 183, 160, 181], calculations have also been carried out assuming $c$ to be a constant. In addition, we have not calculated the dynamic exponent $z$, which is known to be $z = d$ for the short-range interactions [55].

The frequency-dependent conductivity $\sigma(i\omega_n)$ is linear at low frequencies as [162]

$$\sigma(i\omega_n) = \frac{\sigma^*}{1 + |\omega_n|\tau_c}, \quad (3.33)$$

where $\tau_c \sim 1/\omega_c$ is a nonuniversal relaxation time constrained by the ultraviolet cutoff. Eq. (3.33) can be rewritten in the Drude form of conductivity as

$$\sigma(\omega + i\delta) = \frac{\sigma^*}{1 - i\omega\tau_c}, \quad (3.34)$$

with an extrapolation to $\sigma^*$ at zero frequency. In the calculations of the universal conductivity $\sigma^*$ at each $K_c$, we also use Eq. (3.34), but we get smaller value than

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that we present in the above at each $K_c$. This smallness of $\sigma^*$ is analogous to those in Refs. [148, 162, 183], but we don’t provide them here.
CHAPTER 4

INTERMODULATION COEFFICIENT OF AN INHOMOGENEOUS SUPERCONDUCTOR

4.1 Introduction

Ever since the discovery of high-$T_c$ superconductors [15], many workers have attempted to develop practical applications for them. One potential electronic application is as a microstrip resonator. Such a device has been developed by Willemsen et al. [187], using a high-$T_c$ cuprate material. Even though these devices do not work at very high current densities, they are subject to strong nonlinear effects which mix together microwaves of different frequencies. This mixing, known as intermodulation, was studied experimentally and theoretically by Willemsen et al. [187]; theoretical models to explain their measurements were developed by Dahm and Scalapino [38, 39].

In the model of Refs. [38] and [39], the intermodulation is described in terms of an intermodulation current density $J_{IMD}$, which depends on both temperature and the angle between the direction of current flow and the crystal axes. An equivalent quantity was also considered by Yip and Sauls [193] for a $d$-wave superconductor. Because of low-lying quasiparticles, they found that $J_{IMD}$ was much smaller than for a corresponding $s$-wave superconductor and also that it depended on the angle
between the in-plane current density and the $k$-vector of the gap nodes. Several other workers have also studied this intermodulation and the harmonic generation due to nonlinear effects [191, 167, 121].

In all this chapter, the high-$T_c$ cuprate superconductor was assumed to be homogeneous—that is, the CuO$_2$ layer properties were assumed independent of position within the layer. However, recent experimental work has invalidated this picture. Specifically, in optimally oxygen-doped [140, 50], underdoped [79, 107], and slightly overdoped [107] Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (usually called BSCCO-2212), the superconducting energy gap was found to be spatially varying. This result was obtained from scanning tunneling microscopy/spectroscopy (STM/S) images of the superconducting layers. The differential tunneling conductance spectra of the sample were measured and the position-dependent gap $\Delta$ was inferred from measurements of the energy difference between two coherence peaks in the spectra above and below the Fermi level. In the underdoped sample, the gap was found to map into two distinct regions. One (called the $\alpha$ domains) had a gap $\Delta < 50$ meV; the other (denoted the $\beta$ domains) had $\Delta > 50$ meV. The $\alpha$-domains were identified as superconducting because they showed coherence peaks in the tunneling spectra. The $\beta$-regions were found to be nonsuperconducting and were identified as a pseudogap phase [79, 107], with a large gap. It was concluded that the spatially varying superconducting energy gaps do not arise from impurities, but are instead intrinsic properties of the material. Thus underdoped BSCCO-2212 is an intrinsically granular superconductor.

In this chapter, we consider how this inhomogeneity affects the intermodulation in a high-$T_c$ superconductor, such as BSCCO-2212. As implied above, this intermodulation coefficient is actually a nonlinear transport coefficient. In fact, this coefficient
describes the current-dependence of the superfluid density in the superconductor. This current-dependence is particularly strong in the \(d\)-wave high-\(T_c\) materials because quasiparticles are excited even at very low applied currents. We will show that \(J_{\text{IMD}}\) is formally analogous to another coefficient, well known in the study of nonlinear dielectrics. Using this connection, we will demonstrate that \(J_{\text{IMD}}\) is very sensitive to the detailed geometry of the superconducting inhomogeneity.

We will consider only 2D systems and only very low frequencies. This regime is appropriate to the cuprate superconductors, where superconductivity is believed to occur within CuO\(_2\) planes. Our low-frequency approach should be applicable so long as the length scale of the inhomogeneity is much smaller than the wavelength of the applied microwave fields, a condition easily satisfied at microwave frequencies.

The remainder of this chapter is organized as follows. In Sec. 4.2, we present the formalism for calculating the enhancement of \(J_{\text{IMD}}\) in an inhomogeneous 2D superconductor. In Sec. 4.3, we give several analytical results for the relevant enhancements, for \(J_{\text{IMD}}\), and for the effective superfluid densities, based on the analogy to a nonlinear dielectric composite. Section 4.4 presents a concluding discussion.

4.2 Formalism

4.2.1 Intermodulation Coefficients from Ginzburg-Landau Theory

We begin by expressing \(J_{\text{IMD}}\) in terms of coefficients of the Ginzburg-Landau free energy density \(F\). In the absence of a vector potential \(A\), \(F\) takes the standard form

\[
F = a|\psi|^2 + \frac{b}{2}|\psi|^4 + C|\nabla \psi|^2, \tag{4.1}
\]
where $a$, $b$, and $C$ are appropriate constants, and $\psi$ is the complex position-dependent Ginzburg-Landau wave function. For the present problem, we will eventually assume that all three constants are functions of position. We also write $C = \hbar^2/(2m^*)$, where $m^*$ is a quantity with dimensions of mass.

The local supercurrent density takes the usual quantum-mechanical form

$$J = \frac{e^*}{2m^*} \left[ \psi^* (-i\hbar \nabla) \psi + \text{c.c.} \right],$$

(4.2)

where $e^*$ is the charge of a Cooper pair and ‘c.c.’ denotes a complex conjugate. We write $\psi = |\psi| \exp(i\phi)$ and initially assume that $\phi$, but not $|\psi|$, is position-dependent, so that

$$J = \frac{e^*}{m^*} |\psi|^2 (\hbar \nabla \phi).$$

(4.3)

In the limit of very small current density, the wave function is found by minimizing the free energy with respect to $|\psi|^2$, which gives the standard expression $|\psi|^2 = -a/b$. This quantity is positive if $a < 0$.

If there is a finite phase gradient, $F$ takes the form

$$F = a|\psi|^2 + \frac{b}{2} |\psi|^4 + \frac{\hbar^2}{2m^*} |\psi|^2 |\nabla \phi|^2.$$

(4.4)

Minimizing with respect to the modulus of the wave function at fixed $\nabla \phi$, we find that

$$|\psi|^2 = -\frac{a + (\hbar^2/(2m^*)) |\nabla \phi|^2}{b}.$$ 

(4.5)

The corresponding current density takes the form

$$J = -\frac{e^*}{m^*} \left[ \frac{a}{b} (\hbar \nabla \phi) + \frac{\hbar^2}{2m^* b} |\nabla \phi|^2 \hbar \nabla \phi \right].$$

(4.6)

In the above discussion, we have assumed that $|\psi|$ is position-independent, so that $\nabla |\psi| = 0$. Even if $\nabla |\psi| \neq 0$, Eq. (4.3) for $J$ remains valid. However, there is an extra
term in the free energy density; so Eq. (4.5), and hence Eq. (4.6) do not hold exactly. Nevertheless, we shall assume that the most important effects of inhomogeneity can be included by writing

$$J = K_1(x) \nabla \phi \left(1 - K_2(x) |\nabla \phi|^2 \right), \quad (4.7)$$

with appropriate coefficients $K_1(x)$ and $K_2(x)$.

We now show that Eq. (4.6) (for a uniform superconductor) contains the intermodulation phenomenon of interest. First, we rewrite Eq. (4.6) for a uniform superconductor as

$$J = K_1 \nabla \phi (1 - K_2 |\nabla \phi|^2), \quad (4.8)$$

where $K_1$ and $K_2$ are related to the original $a$, $b$, and $m^*$. If $A \neq 0$, in order for the gradient to remain gauge-invariant, we must make the replacement $-i\hbar \nabla \rightarrow -i\hbar \nabla - (e^*/c) A$, or equivalently

$$\nabla \phi \rightarrow \nabla \phi - \frac{e^*}{\hbar c} A. \quad (4.9)$$

Thus, if $A \neq 0$ but the phase is uniform, we must rewrite Eq. (4.8) as

$$J = -\frac{e^* K_1}{\hbar c} A \left(1 - \frac{e^* K_2}{\hbar^2 c^2} |A|^2 \right). \quad (4.10)$$

To order $A^3$ (or $J^3$), we can replace $|A|^2$ on the right-hand side of this expression by $[\hbar c/(e^* K_1)]^2 |J|^2$, which gives

$$J = -\frac{e^* K_1}{\hbar c} A \left(1 - \frac{J^2}{J_{IMD}^2} \right), \quad (4.11)$$

where

$$J_{IMD}^2 = \frac{K_1^2}{K_2}. \quad (4.12)$$

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Finally, we show that Eq. (4.11) implies a current-dependent penetration depth. To see this, we first take the curl of Eq. (4.11) in the low current limit to get

$$\nabla \times \mathbf{J} = -\alpha' \mathbf{B}, \quad (4.13)$$

where $\alpha' = e^* K_1 / (hc)$. When this equation is combined with Ampère’s law, $\nabla \times \mathbf{B} = 4\pi \mathbf{J} / c$, we get $\nabla^2 \mathbf{J} - [1/\lambda^2(T, 0)] \mathbf{J} = 0$, where

$$\frac{1}{\lambda^2(T, 0)} = \frac{4\pi e^* K_1}{hc^2}, \quad (4.14)$$

and $\lambda(T, 0)$ is the zero-current penetration depth at temperature $T$. Thus finally

$$\mathbf{J} = -\frac{c}{4\pi \lambda^2(T, 0)} \mathbf{A} \left( 1 - \frac{J^2}{J_{\text{IMD}}^2} \right). \quad (4.15)$$

Eq. (4.15) has the form $\mathbf{J} = -\mu(T, J) \mathbf{A}$, where $\mu(T, J) = c/[4\pi \lambda^2(T, J)]$, $\lambda(T, J)$ being the temperature- and current-dependent penetration depth. Thus Eq. (4.15) is equivalent to

$$\frac{1}{\lambda^2(T)} = \frac{1}{\lambda^2(T, 0)} \left( 1 - \frac{J^2}{J_{\text{IMD}}^2} \right). \quad (4.16)$$

To order $J^2$, this result is equivalent to Eq. (7) of Ref. [38] ¹.

### 4.2.2 Estimate of Ginzburg-Landau Parameters for Cuprate Superconductors

Within the Ginzburg-Landau formalism, the penetration depth is related to the order parameter $|\psi|$ by

$$\frac{1}{\lambda(T, 0)} = \left( \frac{4\pi e^* |\psi|^2}{m^* c^2} \right)^{1/2}, \quad (4.17)$$

¹This work doesn’t consider the dependence of the coefficient $J_{\text{IMD}}$ on the angle $\theta$ between the current and the $a$-axis of the CuO$_2$ plane, such as would be expected in a superconductor having a $d_{x^2-y^2}$ order parameter.
and \(|\psi|^2 = -a/b\), provided that \(a < 0\). Taking \(a = -\alpha(1 - T/T_{c0})\), where \(\alpha > 0\) and \(T_{c0}\) is the mean-field transition temperature, we obtain

\[ |\psi|^2 = \alpha (1 - T/T_{c0}) / b \]  

(4.18)

and

\[ \frac{1}{\lambda^2(T, 0)} = \frac{4\pi e^2 \alpha (1 - T/T_{c0})}{m^* c^2 b}. \]  

(4.19)

In Eq. (4.18), \(\psi\) has dimensions of a wave function. Lacking an accepted microscopic theory for the high-\(T_c\) cuprates, we may estimate \(\alpha\) and \(b\) using BCS theory, as discussed, for example, by de Gennes [44]. The result is

\[ \alpha = \frac{\hbar^2}{2m^* \xi_0^2} \]  

(4.20)

and

\[ b = \frac{\hbar^4}{4m^* \xi_0^4 N(0)(k_B T_{c0})^2}. \]  

(4.21)

Here, \(N(0)\) is the single-particle density of states at the Fermi energy (measured in states per unit energy per unit volume), and \(\xi_0\) is a temperature-independent coherence length. The penetration depth is then found to be determined by the equation

\[ \frac{1}{\lambda^2(T, 0)} = \frac{32\pi e^2}{\hbar^2 c^2} N(0) \xi_0^2 \Delta^2(T), \]  

(4.22)

where \(\Delta(T)\) is the equilibrium value of the energy gap, given by the equation

\[ \Delta^2(T) = 9.38 k_B^2 T_{c0}(T_{c0} - T). \]  

(4.23)

Now according to experiments [79, 140, 107, 50], in the small gap regions, BSCCO-2212 has a sizable superfluid density, whereas in the large gap regions, the superfluid density is small or zero. If we interpret \(1/\lambda^2(T, 0)\) as proportional to the local superfluid density, then this experimental result implies that \(1/\lambda^2(T, 0)\) should
vary *inversely* with $\Delta(T)$. In order for this to be consistent with Eq. (4.22), the quantity $N(0)\xi_0^4\Delta^2(T)$ should vary inversely with $\Delta(T)$. We therefore assume that $1/\lambda^2(T,0) \propto \Delta^{-x}(T)$, where $x > 0$ is some number which characterizes BSCCO-2212. While this is a highly oversimplified model, it does suggest how $J_{\text{IMD}}$ is influenced by the inhomogeneity of the high-$T_c$ layer.

### 4.3 Model Calculations

#### 4.3.1 Analogy to a Composite Dielectric Medium with a Cubic Nonlinearity

We now apply the above results to calculate $J_{\text{IMD}}$ for several models of inhomogeneous superconducting layers. In all cases, we attempt to choose the layer properties to resemble those reported in experiments on BSCCO-2212. Our goal is to solve Eq. (4.7) for $J(x)$ and $\phi(x)$ for some prescribed inhomogeneous superconductors. We assume that $K_1(x)$ and $K_2(x)$ are specified, but random.

The present problem is *formally* equivalent to a randomly inhomogeneous dielectric with a cubic nonlinearity [197, 168]. In the latter problem, the electric field $\mathbf{E}$ and electric displacement $\mathbf{D}$ are related by

$$
\mathbf{D}(x) = \epsilon(x)\mathbf{E}(x) + \chi(x)|\mathbf{E}(x)|^2\mathbf{E}(x),
$$

$$
\nabla \times \mathbf{E} = 0,
$$

$$
\nabla \cdot \mathbf{D} = 0.
$$

(4.24)

For the intermodulation problem, the analogous equation is Eq. (4.7), supplemented by the steady-state charge conservation condition $\nabla \cdot \mathbf{J} = 0$. Thus $-\nabla \phi$ plays the role of $\mathbf{E}$ in the intermodulation problem, and the phase $\phi$ plays the role of the scalar potential. The quantities $K_1(x)$ and $K_1(x)K_2(x)$ are analogous to the linear dielectric
function $\epsilon(x)$ and cubic nonlinear susceptibility $\chi(x)$. The quantity $-\nabla \phi$ is, of course, curl-free like $E$ in an electrostatic problem. Thus we are again connecting a divergence-free field to a curl-free field. To treat the intermodulation problem, therefore, we can use all the formal results previously derived for an inhomogeneous nonlinear dielectric, which we now briefly review.

For a material described by Eqs. (4.24), two useful coefficients are the *effective linear dielectric function* $\epsilon_e$ and the *effective cubic nonlinear susceptibility* $\chi_e$. These quantities are defined in terms of the space-averaged electric field $\langle E \rangle$ and displacement $\langle D \rangle$ by

$$\langle D \rangle = \epsilon_e \langle E \rangle + \chi_e |\langle E \rangle|^2 \langle E \rangle. \quad (4.25)$$

As shown in Ref. [168], $\chi_e$ can be expressed as an average over the fourth power of the electric field in the associated linear composite. That is, if $E_{\text{lin}}(x)$ is the electric field in a composite described by the linear relation $D(x) = \epsilon(x)E(x)$, then $\chi_e$ is given by

$$\chi_e E_0^4 = \langle \chi(x) |E_{\text{lin}}(x)|^4 \rangle, \quad (4.26)$$

where $E_0$ is the applied electric field. If the composite medium has $n$ components, the $i$th of which has nonlinear susceptibility $\chi_i$, then Eq. (4.26) can be rewritten as

$$\chi_e = \sum_i p_i \chi_i e_i, \quad (4.27)$$

where $p_i$ is the volume fraction of the $i$th component, $e_i$ is an enhancement factor given by

$$e_i = \frac{\langle |E(x)|^4 \rangle_{i,\text{lin}}}{E_0^4}, \quad (4.28)$$

and $\langle \cdots \rangle_{i,\text{lin}}$ means a space-average within the $i$th component in the related linear medium. Thus $e_i$ describes how much the fourth power of the electric field is enhanced in the $i$th component in the linear limit.
The moments $\epsilon_i$ are difficult to compute exactly, except in a few very simple geometries. We have therefore chosen to make a decoupling approximation [169, 196], specified by

$$\langle |\mathbf{E}(\mathbf{x})|^4 \rangle_{i,\text{lin}} \approx \langle |\mathbf{E}(\mathbf{x})|^2 \rangle^2_{i,\text{lin}}. \quad (4.29)$$

Clearly, the decoupling approximation (4.29) will be most accurate if the fluctuations $\langle |\mathbf{E}(\mathbf{x})|^4 \rangle_{i,\text{lin}} - \langle |\mathbf{E}(\mathbf{x})|^2 \rangle^2_{i,\text{lin}}$ within the $i$th component are small compared to $\langle |\mathbf{E}(\mathbf{x})|^4 \rangle_{i,\text{lin}}$ itself [196]. This assumption is most likely to be accurate in geometries such that $|\mathbf{E}(\mathbf{x})|$ is uniform in each of the nonlinear components, but will be less accurate when the fluctuations are large.

If we make the approximation (4.29), we can express $\langle |\mathbf{E}(\mathbf{x})|^2 \rangle_{i,\text{lin}}$ exactly in terms of the effective linear dielectric function $\epsilon_e$ through the relation

$$\frac{p_i \langle |\mathbf{E}(\mathbf{x})|^2 \rangle_{i,\text{lin}}}{E_{0i}^2} = \frac{\partial \epsilon_e}{\partial \epsilon_i} \equiv F_i. \quad (4.30)$$

Here, $\partial \epsilon_e/\partial \epsilon_i$ is the partial derivative of $\epsilon_e$ with respect to $\epsilon_i$, at constant $\epsilon_j$ ($j \neq i$) and constant volume fractions $p_j$. Given a simple analytical approximation for $\epsilon_e$, these derivatives can be easily computed in closed form, thus yielding a simple analytical approximation for $\chi_e$ as

$$\chi_e = \sum_i \chi_i F_i^2/p_i \quad (4.31)$$

with $\epsilon_i = F_i^2/p_i^2$. We will use this approach, combined with the analogy described above, to obtain approximations for the intermodulation coefficient in an inhomogeneous superconducting layer.

In the present chapter, we use two different approximation methods to calculate $\epsilon_e$: the effective-medium approximation (EMA) [24, 106, 16], and the Maxwell-Garnett approximation (MGA) [106, 16]. The EMA is suitable for a binary composite
with symmetrically distributed components, so that neither can be viewed as the inclusion or the host [169]. In this case, if the components are isotropic, \( \epsilon_e \) satisfies the quadratic equation

\[
p_A \frac{\epsilon_A - \epsilon_e}{\epsilon_e + g(\epsilon_A - \epsilon_e)} + (1 - p_A) \frac{\epsilon_B - \epsilon_e}{\epsilon_e + g(\epsilon_B - \epsilon_e)} = 0. \tag{4.32}
\]

Here \( p_A \) is the volume fraction of the component \( A \), \( \epsilon_A \) and \( \epsilon_B \) are the dielectric functions of the components \( A \) and \( B \), respectively, and \( g \) is a ‘depolarization factor’: \( g = 1/2 \) in 2D and \( g = 1/3 \) in 3D. The physically meaningful solution of Eq. (4.32) is the root which varies continuously with \( p_A \), approaches the correct limits at \( p_A = 0 \) and 1, and has a nonnegative imaginary part when \( \epsilon_A \) and \( \epsilon_B \) are complex [169].

The MGA is more suitable to a binary composite where one component can be regarded as a host in which the other is embedded [196]. When the host material is isotropic and linear, the effective dielectric function of the composite takes the form

\[
\epsilon_e = \epsilon_h \left[ 1 + \frac{p_i (\epsilon_i - \epsilon_h)}{(1 - p_i)[\epsilon_h(1 - g) + \epsilon_i g] + p_i \epsilon_h} \right], \tag{4.33}
\]

where \( p_i \) is the volume fraction of the inclusion, \( \epsilon_i \) and \( \epsilon_h \) are the dielectric functions of the inclusion and the host, respectively, and \( g \) is again the depolarization factor.

### 4.3.2 Application to an Inhomogeneous Superconductor

We can readily use the above analogy to compute the effective nonlinear coefficients for an inhomogeneous superconducting layer. We consider a superconducting layer comprised of two “components,” \( A \) and \( B \), with areal fractions \( p_A \) and \( p_B = 1 - p_A \), which have two different energy gaps. The two components are both intrinsic to the given sample, in the sense that they are not caused by the introduction of nonsuperconducting impurities. A realistic sample of BSCCO-2212 probably has
a continuous distribution of gaps, but we make this simplification for computational convenience.

The effective cubic nonlinear coefficient \((K_1K_2)_e\) takes the form

\[
(K_1K_2)_e = (K_1K_2)_A p_A e_A + (K_1K_2)_B p_B e_B.
\]  

(4.34)

\(J_{\text{IMD}}\) in Eq. (4.12) thus becomes

\[
J_{\text{IMD}} = \frac{K_{1e}}{K_{2e}^{1/2}} = \frac{(K_1K_2)_e}{K_{2e}^{3/2}}.
\]  

(4.35)

To apply the present formalism, we need to find suitable \(K_1\) and \(K_2\) values. From Eqs. (4.6) and (4.8), \(K_1 = -(a/b)(\hbar e^*/m^*)\) and \(K_1K_2 = \hbar^3e^*/(2m^*b)\), giving \(K_2 = -\hbar^2/(2m^*a)\). Using \(a = \alpha(t - 1)\), where \(t = T/T_0\), and taking \(\alpha\) and \(b\) from Eqs. (4.19) and (4.20), we find

\[
K_1 = -\frac{a \hbar e^*}{b m^*} = \frac{\hbar e^2}{4\pi e^* \lambda^2(t, 0)} \tag{4.36}
\]

and

\[
K_2 = \frac{\hbar^2}{2m^*|a|} = \xi^2(t). \tag{4.37}
\]

In typical cuprate superconductors, \(\xi(t = 0) \sim 15\) Å and \(\lambda(t = 0, J = 0) \sim 1500\) Å; so

\[
K_2 = (15\) Å)^2. \tag{4.38}
\]

To estimate the values of \(K_1\), we first assume that

\[
\frac{1}{\lambda^2(T, 0)} \propto [\Delta(T)]^{-x}. \tag{4.39}
\]

This assumption embodies the experimental observation that the superfluid density is relatively large in regions where the gap is relatively small. In the model calculations
below, we consider two different values of \( x \), in order to see how this value affects the calculated \( J_{\text{IMD}} \).

Eqs. (4.36) and (4.39) can be combined with experiment to get a rough estimate of \( K_1 \). According to Ref. [140], \( \Delta(T) \) ranges from 25 meV to 65 meV in optimally doped BSCCO-2212 at low \( T \). We assume that the mean value, 45 meV, corresponds to \( \lambda(0,0) = 1500 \) Å. This fixes the proportionality constant in Eq. (4.39). Using this proportionality constant and Eq. (4.36), we get

\[
K_1 = 3.49 \times 10^{11} \left( \frac{\Delta(0)}{45 \text{ meV}} \right)^{-x} \text{ esu} / \text{cm} \cdot \text{s}.
\]  

(4.40)

We first assume that \( x = 1 \). Eq. (4.40) implies that \( \Delta(0) = 25 \text{ meV} \) and 65 meV correspond respectively to \( K_{1A} = 6.28 \times 10^{11} \text{ esu} / (\text{cm} \cdot \text{s}) \) and \( K_{1B} = 2.42 \times 10^{11} \text{ esu} / (\text{cm} \cdot \text{s}) \).

For \( K_{2A} \) and \( K_{2B} \), we have no definite information from experiment. We therefore assume simply that \( K_{2A} = K_{2B} \).

In Figs. 4.1 and 4.2, we show the calculated enhancement factors \( e_A \) and \( e_B \) and the corresponding intermodulation critical current density \( J_{\text{IMD}} \) for these models, as functions of \( p_A \). In Figs. 4.1(a), 4.1(c), and 4.2(a) we use the EMA [Eq. (4.32)], while in Figs. 4.1(b), 4.1(d), and 4.2(b) we use the MGA [Eq. (4.33)], with \( B \) considered as the host. In both cases, we combine these approximations with the decoupling approximation [Eqs. (4.29)–(4.31)], to obtain \( J_{\text{IMD}} \).

Fig. 4.2(a) shows that \( J_{\text{IMD}} \) increases linearly with \( p_A \) in the EMA. As in Eqs. (4.34) and (4.35), \( J_{\text{IMD}} \) has contributions from the nonlinearity of both components. While the enhancement factor \( e_A \) is never larger than unity, \( e_B \) can exceed unity, depending on the value of \( p_A \). Thus the nonlinearity of \( B \) has a larger influence on \( J_{\text{IMD}} \) than that of \( A \). As a result, \( J_{\text{IMD}} \) behaves similarly to \( e_B \), having a larger enhancement for the larger \( K_{1A}/K_{1B} \). The MGA results differ little from the EMA.
Figure 4.1: Enhancement factors $p_A e_A$ and $p_B e_B$ for a 2D inhomogeneous superconductor consisting of a binary composite with two different energy gaps, which have the ratio $K_{1A}/K_{1B}$. The effective-medium approximation (EMA) is used in (a) and (c), while the Maxwell-Garnett approximation (MGA) is used in (b) and (d), assuming that $A$ is surrounded by $B$. We use two different sets of $K_1$’s: $K_{1A} = 6.28 \times 10^{11} \text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 2.42 \times 10^{11} \text{esu}/(\text{cm} \cdot \text{s})$ and $K_{1A} = 3.49 \times 10^{11} \text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 2.42 \times 10^{11} \text{esu}/(\text{cm} \cdot \text{s})$ as indicated in the legend. In this and the following plots of Chapter 4, the unit of $K_{1A}$ and $K_{1B}$ is $10^{11} \text{esu}/(\text{cm} \cdot \text{s})$.

results except for a broad peak around $p_A = 0.9$ for the larger ratio of $K_{1A}/K_{1B}$. This peak results from the shift to higher values of $p_A$ of both $p_B e_B$ and $p_A e_A$, seen in the MGA results of Fig. 4.1.

We can also calculate the effective linear coefficients $K_{1e}$ for these two models. In the 2D EMA, $K_{1e}$ satisfies

$$\frac{K_{1A} - K_{1e}}{K_{1A} + K_{1e}} + (1 - p_A) \frac{K_{1B} - K_{1e}}{K_{1B} + K_{1e}} = 0,$$

(4.41)
while in the 2D MGA with $B$ considered as the host, we get

$$K_{1e} = K_{1B} \left[ 1 + \frac{2p_A(K_{1A} - K_{1B})}{(1-p_A)(K_{1A} + K_{1B}) + 2p_AK_{1B}} \right].$$

(4.42)

The $K_{1e}$’s are proportional to the effective superfluid densities (or the effective inverse-square penetration depths) of these 2D materials in the linear limit of very small applied currents. The values calculated from the EMA and MGA are shown in Figs. 4.3(a) and 4.3(b). Both increase monotonically with increasing areal fraction of the small-gap component $A$. The MGA results differ very little from the EMA results.

In Figs. 4.4, 4.5, and 4.6, we show an analogous set of calculations, but with $x = 3$. We again assume a binary distribution of gaps, using the same gaps as in Figs. 4.1–4.3. Because of the larger $x$, the ratio $K_{1A}/K_{1B}$ is larger than in Figs. 4.1–4.3. For $x = 3$, using the same proportionality constant, we find that the gaps

Figure 4.2: Intermodulation critical supercurrent density $J_{\text{IMD}}$ for the 2D inhomogeneous superconductor shown in Fig. 4.1. The EMA method is used in (a), while the MGA method is used in (b), assuming that $A$ is surrounded by $B$. The two sets of $K_1$’s are the same as in Fig. 4.1. Note $10^{18}$ power scale on the $y$-axis.
Figure 4.3: Effective superfluid density $K_{1e}$ for a 2D composite having the same values of $K_{1A}$ and $K_{1B}$ used in the calculations of Figs. 4.1 and 4.2. (a) EMA method; (b) MGA method, taking B (the component with the smaller superfluid density) as the host.

$\Delta(0) = 65 \text{ meV}$ and $25 \text{ meV}$ now correspond to $K_1 = 1.16 \times 10^{11} \text{ esu/(cm} \cdot \text{s)} \equiv K_{1B}$, and $K_1 = 20.35 \times 10^{11} \text{ esu/(cm} \cdot \text{s)} \equiv K_{1A}$.

For $x = 3$, for the larger ratio of $K_{1A}/K_{1B}$, the enhancement factor $p_{B} e_B$ has clear peaks as a function of the areal fraction $p_A$. This behavior is shown in Figs. 4.4(c) and 4.4(d). The peak occurs at around the percolation threshold of $p_A = 0.5$ in the EMA results, but at around $p_A = 0.95$ in the MGA results. In addition, $p_{B} e_B$ is dramatically larger ($\sim 100$) in the MGA than in the EMA, for the larger ratio of $K_{1A}/K_{1B}$. The EMA results are nearly symmetric about $p_A = 0.5$ while the MGA results are very asymmetric. There is also a large difference between results for the two gap ratios in the MGA results, but a smaller one in the EMA. By contrast, $p_{A} e_A$ is monotonic in either the EMA or the MGA. Since $J_{IMD}$ has two contributions, one from the enhancement of $A$ and the other from the enhancement of $B$, one expects
that the behavior of $J_{\text{IMD}}$ in the MGA results will mirror the enhancement factor $p_B e_B$ because $p_A e_A \ll p_B e_B$ for the larger ratio of $K_{1A}/K_{1B}$.

Fig. 4.5 shows the behavior of $J_{\text{IMD}}$ for the $K_1$’s shown in Fig. 4.4. As expected, and as already found for $x = 1$, $J_{\text{IMD}}$ for $x = 3$ generally follows the trend of $p_B e_B$. In particular, because of the clear peak in $p_B e_B$, the EMA results show a weak broad peak near the percolation threshold $p_c$ in $J_{\text{IMD}}$ for the larger ratio of $K_{1A}/K_{1B}$. The EMA and MGA results differ greatly for the larger ratio of $K_{1A}/K_{1B}$, not only in the shape of the curves but also in the magnitude of $J_{\text{IMD}}$. In this case, the MGA results follow mostly the shape of the curve for $p_B e_B$. Although $J_{\text{IMD}}$ increases monotonically with $p_A$ in the EMA results, it drops sharply above $p_A = 0.95$ for
Figure 4.5: Same as Fig. 4.2, except that $x = 3$; so the two sets of $K_1$'s used are $K_{1A} = 20.35 \times 10^{11} \text{esu/(cm·s)}$, $K_{1B} = 1.16 \times 10^{11} \text{esu/(cm·s)}$ and $K_{1A} = 3.49 \times 10^{11} \text{esu/(cm·s)}$, $K_{1B} = 1.16 \times 10^{11} \text{esu/(cm·s)}$. The full curve is scaled to the right axis while the dashed line to the left in (b).

Figure 4.6: Same as Fig. 4.3 except that $x = 3$, corresponding to a much larger ratio of $K_{1A}/K_{1B}$. 

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the larger ratio of $K_{1A}/K_{1B}$ in the MGA results. Overall, the $x = 3$ case produces a much larger value of $J_{\text{IMD}}$ for the larger ratio of $K_{1A}/K_{1B}$. Thus, for a device requiring a large $J_{\text{IMD}}$, these results suggest that the best results would be obtained using an inhomogeneous superconductor with a large gap difference and a large $x$ in a Maxwell-Garnett geometry.

Fig. 4.6 shows the effective superfluid densities $K_{1e}$ with $x = 3$. For the smaller ratio of $K_{1A}/K_{1B}$, the EMA results differ little from the MGA results as we found previously in Fig. 4.3. However, for the larger ratio of $K_{1A}/K_{1B}$, the two differ significantly.

There is no distinction between the inclusion and the host in the EMA method, but there is in the MGA method. In our MGA results thus far, we have assumed that $A$ (the component with the smaller gap) is the inclusion and that $B$ is the host. We now consider the reverse configuration, where $B$ is surrounded by $A$. Results for this configuration are shown in Figs. 4.7, 4.8, and 4.9. The MGA results with $x = 1$ shown in Figs. 4.7(a), 4.7(b), 4.8(a), and 4.9(a) are very similar to the EMA results in Figs. 4.1(a), 4.1(c), 4.2(a), and 4.3(a), respectively; indeed, the results for $J_{\text{IMD}}$ and $K_{1e}$ are almost identical in the two approximations. For the larger value of $K_{1A}/K_{1B}$, the enhancement factor $p_B e_B$ is smaller in Fig. 4.7(b) than in Fig. 4.1(c). In general, the results for this version of MGA do not show the dramatically large increases in $J_{\text{IMD}}$ seen in Fig. 4.5 for the larger ratio of $K_{1A}/K_{1B}$. In Figs. 4.7(c), 4.7(d), 4.8(b), and 4.9(b) we show the same MGA calculations for $p_A e_A$, $p_B e_B$, $J_{\text{IMD}}$, and $K_{1e}$ but with $x = 3$. The behavior does not differ greatly from that seen in the cases with $x = 1$, except for an increase in the magnitudes of $J_{\text{IMD}}$ and $K_{1e}$ for the larger ratio of $K_{1A}/K_{1B}$.
Figure 4.7: Same as Fig. 4.1, except that the MGA method is used in all cases, assuming that $B$ is surrounded by $A$. The two sets of $K_1$’s are $K_{1A} = 6.28 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 2.42 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$ and $K_{1A} = 3.49 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 2.42 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$ in (a) and (b), while they are $K_{1A} = 20.35 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 1.16 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$ and $K_{1A} = 3.49 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$, $K_{1B} = 1.16 \times 10^{11}\text{esu}/(\text{cm} \cdot \text{s})$ in (c) and (d).

### 4.4 Discussion

In this chapter, we have calculated the intermodulation critical current $J_{\text{IMD}}$ in an inhomogeneous 2D superconductor characterized by a binary distribution of energy gaps. To carry out this calculation, we used an analogy between the effective cubic nonlinear response of an inhomogeneous superconductor and the effective cubic nonlinear susceptibility of a composite dielectric medium. Using this analogy, we can apply the formalism previously developed to treat the nonlinear dielectric composite to the inhomogeneous superconductor. This kind of analogy has never been tried before. We found that the cubic nonlinear response of the superconductor could be
Figure 4.8: Intermodulation critical supercurrent density $J_{IMD}$ for the 2D inhomogeneous superconductor shown in Fig. 4.7. The MGA method is used in (a) and (b), assuming that $B$ is surrounded by $A$. The two sets of $K_1$’s used are the same as in Fig. 4.7. Note $10^{18}$ power scale on the $y$-axis.

Figure 4.9: Effective superfluid density $K_{1e}$ corresponding to Figs. 4.7 and 4.8.
expressed in terms of the cubic response of each “component” (i.e., energy gap) and two enhancement factors, each describing the field and current distribution in the related linear medium.

In order to simplify our calculations, we have assumed that the superconducting layer has a binary distribution of energy gaps, $\Delta_A$ and $\Delta_B$ (with $\Delta_B > \Delta_A$), and we have considered three plausible topologies: “effective-medium” topology ($A$ and $B$ symmetrically distributed), and two “Maxwell-Garnett” topologies ($A$ embedded in $B$ and $B$ embedded in $A$). We have treated all three using a simple nonlinear decoupling approximation.

The results for $J_{IMD}$ are dramatically dependent on the assumed topologies. The EMA and the MGA with $B$ in $A$ give rather similar results for ratios of $\Delta_B/\Delta_A$ close to unity, and only modest enhancements of $J_{IMD}$ at any concentrations of $A$. However, the MGA with $A$ (the component with the smaller gap) embedded in $B$ leads to huge enhancements in $J_{IMD}$ compared to its value in either pure $A$ or pure $B$, provided that the two gap values are sufficiently different and that $x$ is large.

In view of these differences, it is of interest to compare our results with the detailed measurements of Davis et al. [107]. These experiments do not provide results directly for $J_{IMD}$. However, they do provide some hints about a possible connection. In particular, in experiments on as-grown Ni-doped samples, Ni scattering resonances were observed only in the regions where $\Delta < 50$ meV, which were identified as superconducting regions, i.e., regions with superconducting phase coherence. In our results, most of the enhancement in $J_{IMD}$ comes from $K_{1A}$, which corresponds to the
component with a low energy gap. Therefore the regions of enhanced $J_{\text{IMD}}$ correspond to regions of small energy gap, and also regions of enhanced phase coherence according to the measurements of Ref. [107].

A striking feature of our results is the large difference between the EMA results and the MGA results, especially for binary composites with a large $x$ and very different energy gaps. Which of these approximations is the most correct? In fact, there is not a single correct answer for all materials: the correct choice depends on the actual topology of the material of interest. In particular, we do not know, beforehand, whether the energy gaps in an inhomogeneous superconductor are distributed at random throughout the CuO$_2$ planes or whether regions with one magnitude of energy gap are preferentially surrounded by those of the other energy gap. This topology determines whether we should use the EMA or the MGA approach.

In the experimental gap maps [107], the low-$\Delta$ regions are surrounded by the high-$\Delta$ regions in the underdoped BSCCO-2212 sample, but the high-$\Delta$ regions are surrounded by the low-$\Delta$ regions in the slightly overdoped as-grown BSCCO-2212 sample. Therefore, it appears that we can apply the MGA method to both cases, but with the roles of inclusion and host reversed in each case. But even this description of the distribution may be a simplification of the true gap distribution, which is probably continuous, not a discrete binary distribution. Ideally, we should consider such a continuous distribution of energy gaps.

The great sensitivity of $J_{\text{IMD}}$ to composite geometry, as found in the present chapter, is not surprising, in view of earlier work on transport in linear and nonlinear composite conductors and dielectric media. For example, it is well known that the critical exponents describing transport, especially nonlinear transport, in composite

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media are sensitive to features of the local geometry [146, 66, 145, 180]. We speculate that, depending on the precise nature of this geometry, $J_{\text{IMD}}$ either diverges or goes to zero near a percolation threshold according to an appropriate critical exponent.

Finally, we comment briefly on possible device implications of the present results. A useful microstrip resonator would usually have a minimum of intermodulation, or interference between different frequencies [147]. To achieve this, one would probably want a $J_{\text{IMD}}$ which is as large as possible, because this would lead to $1/\lambda^2(T, J)$ which has the weakest dependence on current. Surprisingly, we find here that $J_{\text{IMD}}$ can actually be increased in some inhomogeneous superconductors, provided that the topology is suitable. Thus the inhomogeneity which appears to be unavoidable in the high-$T_c$ cuprates may even be an advantage in constructing useful microwave devices.
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CHAPTER 5

NEW METHOD TO CALCULATE ELECTRICAL FORCES ACTING ON A SPHERE IN AN ELECTRORHEOLOGICAL FLUID

5.1 Introduction

An electrorheological (ER) fluid is a material whose viscosity changes substantially with the application of an electric field [69, 142, 67, 61]. Generally, such fluids are suspensions of spherical inclusions of dielectric constant $\epsilon_i$ in a host fluid of a different dielectric constant $\epsilon_h$. It is believed that the viscosity changes because the spheres acquire electric dipole moments when an electric field is applied, then move under the influence of the induced dipole-dipole interactions. These forces typically cause the spheres to line up in long chains parallel to the applied field, thereby increasing the viscosity of the suspension. The viscosity relaxes to its usual value when the field is turned off, and the chain-like structure disappears.

ER fluids have potential applications as variable viscosity fluids in automobile devices [71], vibration control [164], and elsewhere. Furthermore, their operating principle is also relevant to other materials, such as magnetorheological (MR) fluids [62, 123, 85]. These are suspensions of magnetically permeable spheres in a fluid
of different permeability, whose viscosity can be controlled by an applied magnetic field.

To obtain a quantitative theory of how ER (and MR) fluids work, one needs to understand the force induced between the spheres when an electric field is applied. At large separations, this force is just that between two interacting electric dipoles whose magnitude is that of a single sphere in an external electric field. But at smaller separations, the force deviates from the dipole-dipole form. Besides this electrostatic interaction between the spheres, there are other forces acting on the spheres, including a viscous frictional force from the host fluid, and a hard-sphere force when the two dielectric spheres come in contact. In the present chapter, we will be concerned only with the electrostatic force.

A number of existing theories go beyond the dipole-dipole approximation in calculating electrostatic forces in ER fluids [96, 95, 32, 40, 43, 93, 174, 172, 171, 37], and several experiments have been carried out which are relevant to forces in the nondipole regime (see, e.g., Refs. [186, 185, 184]). Klingenberg et al. [96, 95] have incorporated both multipole and multibody effects into the sphere-sphere interactions, using a perturbation analysis. Chen et al. [32] have described a multipole expansion for the forces acting on one sphere in a chain of spheres in a fluid of different dielectric constant, and find a strong departure from the dipolar limit when the particles are closer than about one diameter. Davis [40] has calculated the electrostatic forces between dielectric spheres in a host fluid directly, using a finite element approach to solve Laplace’s equation for a chain of particles in a host dielectric. In a more recent work [43], he has used an integral equation approach to calculate the interparticle forces in ER fluids, including effects due to time-dependent application of an external
field and nonlinear fluid conductivity. A finite-element approach has also been used by Tao et al. [174] to solve Laplace’s equation and obtain the electrostatic interactions between particles in a chain of dielectric spheres in a host fluid; they found, as in Ref. [40], that the dipole-dipole approximation is reasonably accurate for large separations or moderate dielectric mismatches, but fails in closely spaced particles and large mismatches. Clercx and Bossis [37] have gone beyond the approximation of dipolar interactions to include multipolar and many-body interactions, expressed in terms of the induced multipole moments on each sphere; they also obtain an expression for the forces in terms of these induced multipole moments.

Several authors have included the effects of finite conductivity on forces in electrorheological fluids, and have also considered how such forces depend on frequency. Davis [42] has analyzed polarization forces and related effects of conductivity in ER fluids. Tang et al. [172, 171] have calculated the attractive force between spherical dielectric particles in a conducting film. Khusid and Acrivos [93] have considered electric-field-induced aggregation in ER fluids, including interfacial polarization of the particles, the conductivities of both the particles and the host fluid, and dynamics arising from dielectric relaxation. Claro and Rojas [36] have calculated the frequency-dependent interaction energy of polarizable particles in the presence of an applied laser field within the dipole approximation; they considered primarily optical frequencies rather than the low frequencies more characteristic of ER fluids. Ma et al. [114] have considered several frequency-dependent properties of ER systems, starting from a well-known spectral representation [17, 18, 16] for the dielectric function of a two-component composite medium.
In this chapter, we present a simple method for calculating the electric-field-induced force between two dielectric spheres in a host of a different dielectric constant, at any separation. Our approach is applicable, in principle, to spheres of different sizes, to particles of shape other than spherical, to suspensions in which either the particle or the host, or both, have nonzero conductivities, and to systems in which the constituents have frequency-dependent complex dielectric functions. It can also be used to calculate the electrostatic force on one particle which is part of a many-particle system, and thus is not limited to two-body interactions. It should thus be useful in quite general circumstances.

Our approach is based on a method for calculating the total electrostatic energy of a suspension of (two or more) spheres in a host material of different dielectric constant. This total energy can be expressed in terms of a certain pole spectrum describing the dielectric resonances of the multi-sphere system [17, 18, 16]. This representation has previously been used to calculate the frequency-dependent shear modulus, static yield stress, and structures of certain ER systems [114, 68, 176, 175]. The radial component of the force between the two spheres involves a derivative of this energy with respect to the sphere separation. But rather than evaluating this derivative numerically, as in previous work [114], we express this derivative in closed analytical form in terms of the pole spectrum and certain matrix elements involving these poles. The formula is reminiscent of the Hellmann-Feynman expression for forces in quantum-mechanical systems [76, 53], but differs from it in some important ways.

The remainder of this chapter is organized as follows. In Sec. 5.2, we present the formalism necessary to calculate the forces in a system of two (or more) dielectric spheres in a host medium. In Sec. 5.3, we give several numerical examples of these
forces, at both zero and finite frequencies, for a two-sphere system. Section 5.4 presents a concluding discussion.

5.2 Formalism

Let us assume that we have a composite consisting of inclusions of dielectric constant \( \epsilon_i \) in a host of dielectric constant \( \epsilon_h \), both of which may be complex and frequency-dependent. We will assume that a uniform electric field \( \text{Re} [E_0 e^{-i\omega t}] \) is applied in an arbitrary direction (we take \( E_0 \) real). A static applied field (\( \omega = 0 \)) is a special case of this general form. The suspension is described by a frequency- and position-dependent, but isotropic, complex dielectric function \( \epsilon(x, \omega) \). We also assume that the system is in the “quasistatic regime.” In this regime, the product \( k \xi \ll 1 \), where \( k \) is the wave vector and \( \xi \) is a characteristic length scale describing the spatial variation of \( \epsilon(x, \omega) \). Under these conditions, the local electric field \( E(x, \omega) = -\nabla \Phi \), where \( \Phi \) is the electrostatic potential. The approach which we use automatically includes all local field effects.

Since our force expressions differ slightly at zero and finite frequencies, we will first present the formalism at \( \omega = 0 \), and then generalize the results to finite \( \omega \).

5.2.1 Zero Frequency

If the position of the spheres is fixed, the total electrostatic energy may be written in the form

\[
W = \frac{V}{8\pi} \epsilon_e E_0 \cdot E_0, \tag{5.1}
\]

where \( V \) is the system volume, and \( \epsilon_e \) is the effective dielectric constant. Eq. (5.1) is, in fact, a possible definition of \( \epsilon_e(\omega) \) [16]. To produce this applied field, we require
that \( \Phi(x) = -E_0 \cdot x \) at the boundary \( S \) of the system, which is assumed to be a closed surface enclosing \( V \).

For an isotropic composite, \( \epsilon_c \) may be written in terms of a certain pole spectrum of the composite as \([17, 18, 16]\)

\[
1 - \frac{\epsilon_c}{\epsilon_h} = \sum_{\alpha} \frac{B_{\alpha}}{s - s_{\alpha}}, \tag{5.2}
\]

where

\[
s \equiv \frac{1}{1 - \epsilon_i/\epsilon_h}, \tag{5.3}
\]

\( s_{\alpha} \) is a pole, and \( B_{\alpha} \) the corresponding residue. The poles \( s_{\alpha} \) are confined to the interval \( 0 \leq s_{\alpha} < 1 \).

We now suppose that the system consists of a suspension of spheres of dielectric function \( \epsilon_i(\omega) \) in a host of dielectric function \( \epsilon_h(\omega) \). The sphere centered at \( \mathbf{R} \) is assumed to have radius \( a_\mathbf{R} \). Then the \( s_{\alpha} \)’s satisfy the determinantal condition

\[
\det[s_{\alpha}I - H] = 0, \tag{5.4}
\]

where \( H \) is a certain Hermitian matrix and \( I \) is the identity matrix. It is convenient to express this matrix in terms of a basis indexed by \( \mathbf{R}, \ell, m \), where \( \ell \) and \( m \) are angular momentum indices. In terms of this basis, it is found that \( H \) has the following matrix elements \([18]\):

\[
H_{\mathbf{R}\ell m;\mathbf{R}'\ell' m'} = s_{\ell} \delta_{\ell,\ell'} \delta_{m,m'} \delta_{\mathbf{R},\mathbf{R}'} + Q_{\mathbf{R}\ell m;\mathbf{R}'\ell' m'}(1 - \delta_{\mathbf{R},\mathbf{R}'}), \tag{5.5}
\]

where the variable \( s_{\ell} \) is given by

\[
s_{\ell} = \frac{\ell}{2\ell + 1}. \tag{5.6}
\]
The matrix element $Q_{R^\ell m : R'^\ell m'}$ is given by

\[ Q_{R^\ell m : R'^\ell m'} \equiv (-1)^{\ell' + m'} \frac{a_R^{\ell+1/2}}{|R' - R|^{\ell + \ell' + 1}} \left( \frac{\ell'}{(2\ell + 1)(2\ell' + 1)} \right)^{1/2} \times \left[ (\ell + m)!((\ell' - m)!(\ell' + m')!((\ell' - m')!)^{1/2} \times e^{i\phi_{R' - R}(m' - m)} P_{\ell' + \ell}^{m' - m}(\cos \theta_{R' - R}), \right. \]  

(5.7)

where $\theta_{R' - R}$ and $\phi_{R' - R}$ are polar and azimuthal angles of the vector $R' - R$, and the functions $P_{\ell' + \ell}^{m' - m}$ are the associated Legendre polynomials.

The corresponding residues $B_\alpha$ may be expressed as

\[ B_\alpha = |M_\alpha|^2, \]  

(5.8)

where in general,

\[ M_\alpha = \frac{\sum_{R^\ell m} A_{R^\ell m}^{\alpha} M_{R^\ell m}}{\left( \sum_{R^\ell m} |A_{R^\ell m}^{\alpha}|^2 \right)^{1/2}}, \]  

(5.9)

Here the eigenvalues and eigenfunctions of $H$ are labeled by $\alpha$; the eigenfunctions have components $A_{R^\ell m}^{\alpha}$, where $R$ labels the spheres, and $\ell$ and $m$ denote the angular momentum "quantum numbers" of the corresponding single-sphere eigenfunctions.

Hereafter, we choose $A_{R^\ell m}^{\alpha}$ to be normalized so that

\[ \sum_{R^\ell m} |A_{R^\ell m}^{\alpha}|^2 = 1. \]  

(5.10)

The quantity $M_\alpha$ is proportional to the component of the electric dipole moment of the mode $\alpha$ in the direction parallel to $E_0$. The subscript $\ell = 1, 2, \ldots$, and $-\ell \leq m \leq \ell$.

We choose the polar axis for the spherical harmonics to be the $z$ axis. We first assume that the applied field $E_0$ is also parallel to the $z$ axis. Then $M_{R^\ell m}$ takes the form [18]

\[ M_{R^\ell m} \equiv M_{R^\ell m}^{||} \left( \frac{V_R}{V} \right)^{1/2} \delta_{m,0} \delta_{\ell,1}, \]  

(5.11)
where $v_R$ is the volume of the sphere centered at $R$ and the superscript denotes that the field is parallel to the $z$ axis.

Next, we consider an applied electric field perpendicular to the $z$ axis. If we continue to define the spherical harmonics relative to the $z$ axis, then the $M_{R\ell m}$ now takes the form

$$M_{R\ell m} = M_{R\ell m}^\perp = \left(\frac{v_R}{2V}\right)^{1/2} (\delta_{m,1} + \delta_{m,-1}) \delta_{l,1},$$

(5.12)

where the superscript denotes $E_0 \perp \hat{z}$.

We now apply the above formalism to calculate the force between two spheres of dielectric constant $\epsilon_i$ in a host of dielectric constant $\epsilon_h$, as a function of their separation and the direction of the applied electric field. For convenience, we continue to choose the axis joining the spheres to be parallel to the $z$ axis. We consider two directions for the applied electric field: $E_0 = (0, 0, E_0)$ (“parallel configuration”) and $E_0 = (E_0, 0, 0)$ (“perpendicular configuration”). The geometry is shown in Fig. 5.1. To obtain the force, we first calculate the total energy of a pair of spheres in a host as a function of their separation. The force on either sphere can then be computed as the gradient of this total energy with respect to separation. This derivative can be computed analytically in a simple way, as we now show.

We shall obtain this force on the $R$th sphere for an arbitrary assembly of spheres, then specialize to two spheres. To do this, it is convenient to rewrite the residues $B_\alpha$ in “bra-ket” notation. Such a description is possible because we are dealing with the eigenstates of a Hermitian operator, namely $H$. We therefore introduce a normalized ket

$$|0\rangle \equiv \sum_{R\ell m} |R\ell m\rangle \langle R\ell m|0\rangle.$$  

(5.13)
Figure 5.1: Geometry considered in most of our calculations: Two identical spheres of radius \( a \) are located at the origin and at \( z = R \), and are contained in a host material. \( \delta \) is the surface-to-surface distance between the two spheres. The complex dielectric function of the spheres is \( \epsilon_i(\omega) \) and that of the host material is \( \epsilon_h(\omega) \). A spatially uniform electric field \( \text{Re} \left[ E_0 e^{-i\omega t} \right] \) is applied in the \( z \) direction in (a) and in the \( x \) direction in (b).

|0\rangle is thus expressed as a column vector in the \( |R\ell m\rangle \) basis. The components of \( |0\rangle \) in that basis are denoted as \( \langle R\ell m|0 \rangle \).

In the parallel configuration, we take

\[
\langle 0| R\ell m \rangle = \langle R\ell m|0 \rangle = \left( \frac{v_R}{v_{\text{tot}}} \right)^{1/2} \delta_{\ell,1} \delta_{m,0}, \tag{5.14}
\]

where

\[
v_{\text{tot}} = \sum_R v_R \tag{5.15}
\]

is the total volume of inclusions. With this choice of factor multiplying the delta functions, the ket \( |0\rangle \) satisfies the normalization condition

\[
\langle 0|0 \rangle = \sum_{R\ell m} \langle 0| R\ell m \rangle \langle R\ell m|0 \rangle = \sum_{R\ell m} |\langle R\ell m|0 \rangle|^2 = 1. \tag{5.16}
\]

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In the perpendicular configuration,

\[ \langle 0|R\ell m \rangle = \langle R\ell m|0 \rangle = \left( \frac{v_R}{2v_{\text{tot}}} \right)^{1/2} \delta_{\ell,1}(\delta_{m,1} + \delta_{m,-1}) \]  

and again the coefficients are chosen so that the normalization condition (5.16) is valid.

We also introduce a normalized ket \(|\alpha\rangle\) with coefficients

\[ \langle R\ell m|\alpha \rangle \equiv A^{\alpha}_{R\ell m}. \]  

With this choice and Eq. (5.10), \(|\alpha\rangle\) satisfies the normalization condition

\[ \langle \alpha|\alpha \rangle \equiv \sum_{R\ell m} |\langle R\ell m|\alpha \rangle|^2 = 1. \]  

In this notation, the matrix element \(M_\alpha\) satisfies

\[ M_\alpha = \left( \frac{v_{\text{tot}}}{V} \right)^{1/2} \langle 0|\alpha \rangle = \left( \frac{v_{\text{tot}}}{V} \right)^{1/2} \sum_{R\ell m} \langle 0|R\ell m \rangle \langle R\ell m|\alpha \rangle = \left( \frac{v_{\text{tot}}}{V} \right)^{1/2} \sum_{R\ell m} \langle 0|R\ell m \rangle A^{\alpha}_{R\ell m}. \]  

With these definitions and using Eqs. (5.8), (5.9), (5.16), and (5.19), Eq. (5.2) can be rewritten as

\[ 1 - \frac{\epsilon_e}{\epsilon_h} = \frac{v_{\text{tot}}}{V} \sum_{\alpha} \frac{\langle \alpha|\alpha \rangle}{s - s_\alpha}. \]  

The explicit forms of \(M_\alpha\) are given by Eqs. (5.9) and either (5.11) or (5.12).

To obtain the forces, we rewrite Eq. (5.21) as

\[ 1 - \frac{\epsilon_e}{\epsilon_h} = \frac{v_{\text{tot}}}{V} \langle 0|G(s)|0 \rangle, \]  

where

\[ G(s) \equiv \sum_{\alpha} \frac{|\alpha\rangle\langle \alpha|}{s - s_\alpha} = (sI - H)^{-1} \]  

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is a Green’s function for this problem and the matrix elements of \( H \) are given by Eq. (5.5). Eq. (5.23) is valid because \( H \) is a Hermitian operator, and hence its eigenvalues, which satisfy the “Schrödinger equation”

\[
H |\alpha\rangle = s_\alpha |\alpha\rangle ,
\]

are real and the corresponding eigenvectors \( |\alpha\rangle \) can be constructed to be orthonormal.

Now, suppose that \( H \) depends on some parameter \( \lambda \). In calculating a force, we will typically choose \( \lambda \) to be one of the Cartesian position coordinates of one of the particles. As shown below, the force will be proportional to the derivative \( \partial \varepsilon_e / \partial \lambda \).

But from Eqs. (5.22) and (5.23), we can write

\[
- \frac{1}{\epsilon_h} \frac{\partial \varepsilon_e}{\partial \lambda} = \frac{\nu_{\text{tot}}}{V} \frac{\partial}{\partial \lambda} \langle 0 | G(s, \lambda) | 0 \rangle = \frac{\nu_{\text{tot}}}{V} \left( 0 | \frac{\partial G(s, \lambda)}{\partial \lambda} | 0 \right),
\]

where the last equality holds because, according to Eqs. (5.14) and (5.17), \( \langle 0 \rangle \) and \( | 0 \rangle \) do not depend on \( \lambda \).

We now show how to calculate \( \partial G(s, \lambda) / \partial \lambda \) explicitly. If we introduce \( U = \partial H / \partial \lambda \), we can write

\[
\frac{\partial G(s, \lambda)}{\partial \lambda} = \lim_{\delta \lambda \to 0} \left[ \{ s I - H(\lambda) \} - U \delta \lambda \right]^{-1} - \left[ s I - H(\lambda) \right]^{-1} \\
= \lim_{\delta \lambda \to 0} \left[ \{ I - (s I - H(\lambda))^{-1} U \delta \lambda \}^{-1} (s I - H(\lambda))^{-1} - \left[ s I - H(\lambda) \right]^{-1} \right] / \partial \lambda \\
\simeq [s I - H(\lambda)]^{-1} U [s I - H(\lambda)]^{-1} \\
= G(s, \lambda) U G(s, \lambda),
\]

plus terms of order \((\partial \lambda)^2\). Hence,

\[
- \frac{1}{\epsilon_h} \frac{\partial \varepsilon_e}{\partial \lambda} = \frac{\nu_{\text{tot}}}{V} \langle 0 | G(s, \lambda) U G(s, \lambda) | 0 \rangle.
\]
Using the representation (5.23) for $G(s, \lambda)$ [and taking the eigenvalue $s_\alpha$ and the state $|\alpha\rangle$ to refer to the Hamiltonian $H(\lambda)$], we can rewrite Eq. (5.27) as

$$-\frac{1}{\epsilon_h} \frac{\partial e}{\partial \lambda} = \frac{v_{\text{tot}}}{V} \sum_\alpha \sum_\beta \frac{\langle 0|\alpha\rangle \langle \alpha|U|\beta\rangle \langle \beta|0\rangle}{(s - s_\alpha)(s - s_\beta)}.$$  \hspace{1cm} (5.28)

Eq. (5.28) is our central formal result.

Eq. (5.25) bears a resemblance to the Hellmann-Feynman theorem in quantum mechanics [76, 53]: in both cases, the derivative of a Hamiltonian with respect to a parameter appears inside a matrix element. But there is an important difference between the two. In the Hellmann-Feynman case, the ket which plays the role of $|0\rangle$ is an eigenstate of the Hamiltonian. Although the ket in that case depends on $\lambda$, the derivative can still be moved inside the bra and ket because the eigenstates are orthonormalized. Here, by contrast, $|0\rangle$ is not an eigenstate of $H$, but it does not depend on $\lambda$; so the derivative can still be moved inside the matrix element.

To show how this form can actually be used to compute the force explicitly, consider just a two-sphere case, the two spheres being located at $(0, 0, 0)$ and $(0, 0, R_0)$. The total energy is given by Eq. (5.1). We consider two configurations for the electric field: $\mathbf{E}_0 = (0, 0, E_0)$ (“parallel configuration”) and $\mathbf{E}_0 = (E_0, 0, 0)$ (“perpendicular configuration”). In both cases, the component of the force on the sphere at $R_0$ along the axis joining the two spheres can be calculated using Eq. (5.28) and the relation

$$F = +\left(\frac{\partial W}{\partial R_0}\right)_\Phi.$$  \hspace{1cm} (5.29)

The positive sign is correct here because the system is held at fixed potential on the boundaries [82]. A positive value of $F$ means that the force is repulsive. Given Eqs.
(5.28) and (5.29), the total radial component of the force is

\[
F = \frac{v_{\text{tot}}|E_0|^2\epsilon_h}{8\pi} \left\langle 0 \left| \frac{\partial G}{\partial R_0} \right| 0 \right\rangle \\
= -\frac{v_{\text{tot}}|E_0|^2\epsilon_h}{8\pi} \sum_\alpha \sum_\beta \frac{\langle 0|\alpha\rangle\langle \alpha|U|\beta\rangle\langle \beta|0 \rangle}{(s-s_\alpha)(s-s_\beta)}. \quad (5.30)
\]

To compute the force explicitly, we have to consider how the “Hamiltonian” \(H\) changes with the separation \(R_0\) of the spheres, so that we can compute the matrix elements of \(U\). The diagonal matrix elements of \(H\) are independent of \(R_0\), while each of the off-diagonal matrix elements, according to Eq. (5.7), consists of a power series in \(1/|R-R'| = 1/R_0\). Hence \(U = \partial H/\partial R_0\) is easily calculated in a closed form.

For the case of two spheres, it is straightforward to calculate this derivative. The eigenstates \(|\alpha\rangle\), as well as \(s_\alpha\), are already known if the original eigenvalue problem involving \(H(R_0)\) has been solved. The state \(|0\rangle\) is given by Eq. (5.14) or (5.17). Therefore, it is straightforward to calculate the matrix elements \(\langle \alpha|U|\beta\rangle\) and hence the force, using Eq. (5.30).

### 5.2.2 Finite Frequencies

The results of the previous subsection are readily generalized to finite frequencies. In this case, the total electrostatic energy will be a sinusoidally varying function of time. The quantities of experimental interest will be the time-averaged electrostatic energy \(W_{\text{av}}\) and time-averaged forces. \(W_{\text{av}}\) is given by the generalization of Eq. (5.1), with an extra factor of \(1/2\) to take into account time-averaging, namely

\[
W_{\text{av}} = \frac{V}{16\pi} \text{Re} [\epsilon_\omega(E_0 \cdot E_0)]. \quad (5.31)
\]

Here the applied field is assumed to be \(E_0 \cos(\omega t) = \text{Re} [E_0 e^{-i\omega t}]\) and \(\epsilon_\omega(\omega)\) is the complex frequency-dependent effective dielectric function. All the remaining equations in
Sec. 5.2.1 continue to be valid up to Eq. (5.29), which is replaced with

\[ F_{av} = \left( \frac{\partial W_{av}}{\partial R_0} \right)_\phi \].

(5.32)

The generalization of Eq. (5.30) is

\[ F_{av} = -\text{Re} \left[ \frac{v_{tot}|E_0|^2 \epsilon_h}{16\pi} \sum_\alpha \sum_\beta \langle 0|\alpha \rangle \langle \alpha |U|\beta \rangle \langle \beta |0 \rangle \frac{(s - s_\alpha)(s - s_\beta)}{(s - s_\alpha)(s - s_\beta)} \right] \].

(5.33)

Expression (5.33) can be evaluated just as at \( \omega = 0 \) and thus the time-averaged force at finite frequency can also be computed explicitly.

### 5.3 Numerical Results

We have applied the above formalism to two spheres of dielectric constant \( \epsilon_1 \) in a host of dielectric constant \( \epsilon_h \). We choose a coordinate system such that the two spheres are located at the origin and at \( \mathbf{R} = R\mathbf{z} \), and we consider two configurations for the applied electric field, \( \mathbf{E}_0 = E_0\mathbf{z} \) and \( \mathbf{E}_0 = E_0\mathbf{x} \), as shown in Fig. 5.1.

Once the elements of the \( H \) and \( U \) matrices are known, the calculation of the interparticle force reduces to an eigenvalue problem. To carry out the various required matrix and vector operations, we used GNU Scientific Library (GSL) routines [58] and C++ complex class library. In the parallel configuration, we calculated all the terms in the \( H \) and \( U \) matrices up to \( \ell_{\max} = 80 \); it is easy to include such a large cutoff because only \( m = 0 \) needs to be considered for this geometry, the polar and azimuthal angles of \( \mathbf{R} \) equaling zero. Despite the large cutoff, most of the contributions to these matrices came from \( \ell < 10 \). Based on this information, we set \( \ell_{\max} = 10 \) for the \( H \) and \( U \) matrices in the perpendicular geometry. Even with this cutoff, the matrices involved in this calculation are large since \( m \) can be nonzero in the perpendicular case: the dimension of the matrix for \( \ell_{\max} = 10 \) is \( 2 \sum_{\ell=1}^{10} (2\ell + 1) = 240 \).
The $H$ matrix for both cases consists of four square blocks. The two diagonal square blocks have diagonal elements $s_{\ell} = \ell/(2\ell + 1)$ with all off-diagonal elements vanishing. The other two (off-diagonal) square blocks have elements $Q_{0\ell m \ell' m'}$. For the $U$ matrix, the diagonal square blocks have all zero elements and the elements of the two off-diagonal blocks equal $\partial Q_{0\ell m \ell' m'}/\partial R$. Once we have calculated all the eigenvalues and eigenvectors of the $H$ matrix, we can compute the $M$, $B$, and hence the force on the sphere from the expectation value of the $U$ matrix, using Eq. (5.30) or (5.33).

As a first example, we have considered $\epsilon_i = 10^5$, $\epsilon_h = 1$. The choice for $\epsilon_i$ approximates the value $\epsilon_i = \infty$ corresponding to two metallic spheres at zero frequency in an insulating host with unit dielectric constant. In Fig. 5.2, we show the magnitude of the calculated radial component of the force between the spheres, as a function of their separation, for both parallel and perpendicular configurations. The force is attractive in the parallel configuration, repulsive in the perpendicular configuration. We have arbitrarily chosen sphere radii of $a = 3.15 \text{ mm}$ and a field strength of $E_0 = 25.2 \text{ V/mm}$ as in recent experiments carried out in Refs. [185, 184] (for different materials). However, the forces are easily scaled with both field strength and sphere radii: for fixed $\epsilon_i$ and $\epsilon_h$ the appropriate scaling relation is

$$F_{12}^{\perp,||} = a^6 E_0^2 f_{\perp,||}(\epsilon_i, \epsilon_h, R/a),$$

where $f_{\perp}$ and $f_{||}$ are functions of $\epsilon_i$, $\epsilon_h$ and the ratio $R/a$.

It is of interest to compare these plots with the behavior expected from an electric dipole-dipole interaction. For two dipoles $\mathbf{p}_1$ and $\mathbf{p}_2$ separated by a distance $R$, this
interaction energy has the well-known form

\[ W_{12} = \frac{\mathbf{p}_1 \cdot \mathbf{p}_2 - 3(\mathbf{p}_1 \cdot \hat{n})(\mathbf{p}_2 \cdot \hat{n})}{R^3}, \]  

(5.35)

where \( \hat{n} \) is a unit vector along the axis joining the two dipoles. In the present case, we have

\[ \mathbf{p}_1 = \mathbf{p}_2 = a^3 \mathbf{E}_0 \frac{\epsilon_1 - 1}{\epsilon_1 + 2}, \]

(5.36)

and thus

\[ W_{12} = a^6 \mathbf{E}_0^2 \left( \frac{\epsilon_1 - 1}{\epsilon_1 + 2} \right)^2 \frac{1 - 3(\mathbf{E}_0 \cdot \hat{n})^2}{R^3}, \]

(5.37)
where $\hat{E}_0$ is a unit vector along the applied electric field. For $\hat{E}_0 \perp \hat{n}$, this reduces to

$$U_{12}^{\perp} = a^6 E_0^2 \left( \frac{\epsilon_i - 1}{\epsilon_i + 2} \right)^2 \frac{1}{R^3}.$$  \hspace{1cm} (5.38)

while for $\hat{E}_0 \parallel \hat{n}$, it becomes

$$U_{12}^{\parallel} = a^6 E_0^2 \left( \frac{\epsilon_i - 1}{\epsilon_i + 2} \right)^2 \frac{1}{R^3}.$$  \hspace{1cm} (5.39)

The corresponding radial components of the forces in the parallel and perpendicular cases are the negative derivatives of these functions with respect to $R$:

$$F_{12}^{\perp} = -\frac{1}{R} F_{12}^{\parallel} = 3 a^6 E_0^2 \left( \frac{\epsilon_i - 1}{\epsilon_i + 2} \right)^2 \frac{1}{R^2}.$$  \hspace{1cm} (5.40)

The values of $F_{12}^{\perp}$ and $F_{12}^{\parallel}$ shown in Fig. 5.2 agree very well with those calculated from Eq. (5.40) (taking $\epsilon_i = \infty$) at large separation ($R \gg a$) but depart strongly at small separation ($R - 2a \ll a$). Once again, a positive sign denotes a repulsive force, while a negative sign denotes an attractive force. For example, the ratio of the two forces in the parallel and perpendicular configurations at small separation has a magnitude greater than 50 for $R = 0.632 \text{ cm} = 2a + 0.002 \text{ cm}$, much larger than the factor of 2 expected from the dipole-dipole interaction. Further examples of this ratio are given in Table 5.1 for various separations.

In Fig. 5.3, we show the corresponding forces in both the parallel and perpendicular configurations for two spheres of different radii, $2a$ and $a/2$, but still for the same dielectric constants as in Fig. 5.2 ($\epsilon_i = 10^5$, $\epsilon_h = 1$). In this case, the ratio of forces in the parallel and perpendicular configurations departs more dramatically from the factor of two at close range than in the case of equal size spheres.

In Figs. 5.4 and 5.5, we test the effect of different inclusion dielectric constants, by calculating the force between two identical spheres, each of radius $a$ and dielectric...
Table 5.1: The ratios of the magnitudes of the forces between two identical spheres in the parallel and perpendicular configurations, calculated at several small separations and assuming $\epsilon_i = 10^5$, $\epsilon_h = 1$, $a = 3.15$ mm, $E_0 = 25.2$ V/mm, and $\omega = 0$. The force is attractive in the parallel configuration, repulsive in the perpendicular configuration.

<table>
<thead>
<tr>
<th>$R$(cm)</th>
<th>Force ratio</th>
<th>$(R-2a)/(2a)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.630</td>
<td>602.3</td>
<td>0.0000</td>
</tr>
<tr>
<td>0.631</td>
<td>88.5</td>
<td>0.0016</td>
</tr>
<tr>
<td>0.632</td>
<td>52.5</td>
<td>0.0032</td>
</tr>
<tr>
<td>0.633</td>
<td>38.8</td>
<td>0.0048</td>
</tr>
<tr>
<td>0.634</td>
<td>31.5</td>
<td>0.0063</td>
</tr>
<tr>
<td>0.635</td>
<td>26.8</td>
<td>0.0079</td>
</tr>
<tr>
<td>0.636</td>
<td>23.5</td>
<td>0.0095</td>
</tr>
<tr>
<td>0.637</td>
<td>21.1</td>
<td>0.0111</td>
</tr>
<tr>
<td>0.638</td>
<td>19.2</td>
<td>0.0127</td>
</tr>
<tr>
<td>0.639</td>
<td>17.7</td>
<td>0.0143</td>
</tr>
<tr>
<td>0.640</td>
<td>16.5</td>
<td>0.0159</td>
</tr>
</tbody>
</table>

Figure 5.3: Same as Fig. 5.2, but for two spheres with radii $2a = 6.30$ mm and $a/2 = 1.575$ mm, respectively. $\delta$ is the separation between sphere surfaces.
constant $\epsilon_i$, in a host of dielectric constant $\epsilon_h = 1$. We plot the radial component of this force, for both the parallel and perpendicular configurations, as a function of $\epsilon_i$, for two different separations between the spheres: $R = 2a + 0.01\,\text{mm}$ and $R = 2a + 10.00\,\text{mm}$, where we again use $a = 3.15\,\text{mm}$. In the second case, the forces are very close to the dipole-dipole predictions. In the first case, the forces exhibit a large departure from the predictions of the dipole-dipole interaction, and this departure becomes greater as $\epsilon_i$ deviates more and more from unity.

![Graph](image.png)

Figure 5.4: The radial component of the force at zero frequency between two identical spheres of dielectric constant $\epsilon_i$, radius $a = 3.15\,\text{mm}$, in a host of dielectric constant $\epsilon_h = 1$, at an intersphere spacing (surface-to-surface separation) of 0.01 mm, plotted as a function of $\epsilon_i$, for (a) electric field parallel to the axis between spheres, and (b) field perpendicular to that axis. We assume an electric field of strength 25.2 V/mm. Negative and positive forces denote attractive and repulsive forces, respectively.

Next, we consider an example in which the dielectric functions of both the inclusion and the host depend on frequency. Specifically, we choose

$$\epsilon_i = \epsilon_{i0} + \frac{4\pi\sigma_i}{\omega}$$

(5.41)
and
\[ \epsilon_h = \epsilon_{h0} + i \frac{4\pi \sigma_h}{\omega}, \]  
(5.42)

where \( i \) is the imaginary unit, \( \epsilon_{i0} \) and \( \epsilon_{h0} \) are the dielectric constants of the inclusion and the host, and \( \sigma_i \) and \( \sigma_h \) are their conductivities, assumed frequency-independent.

The time-averaged forces are now calculated from the generalization of Eq. (5.30) to finite frequencies, namely, Eq. (5.33).

We assume parameters given by Refs. [185, 184], in a recent experimental study. These are listed in Table 5.2. In most cases, the two spherical inclusions are identical, with a dielectric constant and conductivity characteristic of SrTiO\(_3\). For the host fluid, we have considered the various materials used in the measurements of Refs. [185, 184]. (In practice, the nonzero conductivity of SrTiO\(_3\) has negligible effect on force; we have checked this by recalculating the forces with the conductivity set equal to zero and obtained the same results.)
Table 5.2: Parameters for the calculations shown in Figs. 5.6–5.9. The columns denote the material, the real part of its dielectric constant, and its conductivity (in S/m). All except for SrTiO$_3$ are used as host materials in the suspensions.

In Fig. 5.6(a) and 5.6(b), we show the radial component of the calculated time-averaged force on a sphere of SrTiO$_3$ at $R$ in the parallel and perpendicular geometries, for the host materials of silicone oil and castor oil. In both cases, we assume spheres of radius $a = 3.15$ mm, intersphere spacing $\delta = 0.01$ mm, and applied electric field $E_0 = 25.2$ V/mm, as in Refs. [185, 184]. The magnitude of force decreases with increasing frequency, but rapidly converges to a constant value in both cases. The sign of the force is negative in 5.6(a), corresponding to an attractive force, and positive (repulsive) in 5.6(b).

If these were strictly dipole-dipole forces, the time-averaged force on the sphere at $R$ would be given by the generalization of Eq. (5.40) to complex dielectric functions and $\epsilon_h \neq 1$, namely

$$F_{\text{av},12}^{\perp} = -\frac{1}{2} F_{\text{av},12}^{\parallel} = \frac{3}{2R^4} a^6 E_0^2 \Re \left[ \frac{\epsilon_i - \epsilon_h}{\epsilon_i + 2\epsilon_h} \right]^2 \epsilon_h .$$

Thus, in particular, the magnitude of the force in the parallel case would be twice as large as that in the perpendicular case, as in our previous examples. However, in Fig.
Figure 5.6: The radial component of the time-averaged force between two identical spheres of SrTiO$_3$, plotted as a function of frequency for the host materials of silicone oil and castor oil, respectively. For both cases we use $\delta = 0.01$ mm, $a = 3.15$ mm, and $E_0 = 25.2$ V/mm. The electric field is parallel to the line connecting the spheres in (a) and perpendicular to that line in (b). A negative value denotes an attractive force.

5.6, this force ratio is about 50. This difference occurs, as in Figs. 5.2 and 5.3, because of the very small separation ($\delta = 0.01$ mm), which corresponds to a very short-ranged interaction. In the long range limit ($R \gg a$), our calculated magnitude ratio agrees well with the dipole-dipole prediction, as discussed further below. This short-distance deviation from dipole-dipole forces is similar to that seen in Figs. 5.2–5.4.

Fig. 5.6 also shows that there is a substantial difference between the forces for silicone oil and castor oil hosts. This difference is due almost entirely to the difference in the static dielectric constants of these two hosts: the effect of the finite conductivity disappears by about 10 Hz in both cases, whereas the difference between the forces persists to much higher frequencies.
The calculated time-averaged force between spheres of SrTiO$_3$ in a silicone oil host is plotted versus separation in Fig. 5.7 at a frequency of 50 Hz. In order to see the effects of a finite host conductivity, we include this conductivity in Figs. 5.7(a) and 5.7(b) but not in 5.7(c) or 5.7(d). We also set the conductivity of the sphere equal to zero in 5.7(c) and 5.7(d). Clearly, the host conductivity has very little influence on the forces at this frequency. For comparison, we also show the forces as calculated in the dipole-dipole approximation. As can be seen, there is very little difference between the two except for $R < \sim 1.5$ cm. Even at such small spacings, the deviation from the dipole-dipole force is much larger for the parallel than the perpendicular configuration. At a spacing of 0.01 mm, the calculated ratio of force magnitudes in the parallel and perpendicular configurations exceeds a factor of 100.

At sufficiently high host conductivity, our model predicts that the force between spheres changes sign as a function of frequency. This trend is shown in Fig. 5.8 for a separation of $\delta = 0.01$ mm between spheres. The host materials used here are ethyl benzoate, ethyl salicylate, and methyl salicylate, all of which have much greater conductivities than silicone oil. The sign change is due mainly to the greater conductivities, not the differences in static dielectric constants. To check this point, we recalculated the points of Fig. 5.8 assuming the same value of the real part of the dielectric constant for all three host materials; we found that the time-averaged forces changed sign at the same frequencies as in Fig. 5.8. Mathematically, the origin of the sign change is, of course, the dependence of the variable $s$ in Eqs. (5.3) and (5.33) on the host conductivity.

The time-averaged force for this separation ranges from about 1.5 to $-1.5$ dyn for the parallel case, depending on the frequency, and from about $+0.5$ to $-3.0$ dyn for
Figure 5.7: (a) and (b) : Magnitude of the radial component of the time-averaged force between two identical spheres of SrTiO$_3$, divided by $E_0^2$. Also plotted is the corresponding quantity in the dipole-dipole approximation. Both are plotted on logarithmic scale as a function of separation $R$ for a host material of silicone oil and a fixed frequency of 50 Hz. The spheres have radii 3.15 mm. The electric field is parallel to the line between the two spheres in (a) and perpendicular to that line in (b). (c) and (d) : Same as (a) and (b) except that the conductivities of the spheres and the host are set equal to zero in these calculations. The forces are attractive in (a) and (c), repulsive in (b) and (d).

the perpendicular case. At high frequencies, the force approaches $-1.0$ dyn for the parallel case, whatever the host fluid is, and approaches a much smaller magnitude in the perpendicular case. The ratio of these forces differs greatly from the predictions of the dipole-dipole interaction, as expected for such a small separation. At very low frequencies, however, the force ratio appears to approach the dipole-dipole prediction.

Fig. 5.9 shows the frequency dependence of the time-averaged force between two spheres of SrTiO$_3$ for silicone oil and $N_2$ hosts. Both the spacings $\delta$ between the two spheres and the electric field $E_0$ are larger than those for Fig. 5.6; they are given in
Figure 5.8: The radial component of the time-averaged force between two identical spheres of SrTiO$_3$ separated by $R$, plotted as a function of frequency for the host materials of ethyl benzoate, ethyl salicylate, and methyl salicylate, respectively. The electric field is parallel to the line connecting the two spheres in (a) and perpendicular to that line in (b). In all cases, $\delta = 0.01$ mm, $a = 3.15$ mm, and $E_0 = 25.2$ V/mm.

The legends of each figure. We chose these values for the parameters in order to allow comparison with the measurements of Refs. [185, 184]. Evidently, the force between the two spheres is stronger when the two spheres are immersed in a liquid host than in a gas, all the other parameters of the forces being held constant. This behavior can be understood even in the dipole-dipole limit: it is due to the dependence of the force on $\epsilon_h$ as in Eq. (5.43). Also, the low-frequency forces in Figs. 5.9(a) and 5.9(b) and especially 5.9(c) and 5.9(d) depend more weakly on frequency than those in Fig. 5.6. Another point is that, even though the intersphere spacing $\delta$ has been increased to 0.10 and 0.30 mm in these calculations, the calculated forces are still far from the dipole-dipole limit. Specifically, the ratio of the force magnitudes in the parallel and
perpendicular geometries greatly exceeds the factor of two expected in the dipole-dipole limit. However, this ratio is smaller than that of Fig. 5.6, presumably because the intersphere separations are larger than in that figure. Moreover, 5.9(b) even shows that the larger separation has the larger magnitude of force in silicone oil when the electric field is perpendicular to the line between two spheres.

Figure 5.9: The radial component of the time-averaged force between two identical spheres of SrTiO$_3$ separated by $R$, plotted as a function of frequency for host materials consisting of silicone oil [(a) and (b)] and $N_2$ [(c) and (d)], with gap spacings $\delta = 0.10$ mm and $\delta = 0.30$ mm. The applied electric field is $E_0 = 71.3$ V/mm and $a = 3.15$ mm for all the cases. The electric field is parallel to the line between two spheres in (a) and (c), and perpendicular to that line in (b) and (d).

Fig. 5.10 shows the time-averaged force between two identical spheres as a function of frequency $\omega/\omega_p$, where $\omega_p$ is the plasma frequency, for different $\ell_{\text{max}}$. It is assumed that $\epsilon_i$ follows the Drude model with $\omega_p\tau = 20$ and that the host material has $\epsilon_h = 1$. This result validates our assumptions of $\ell_{\text{max}} = 80$ for the parallel configuration and
\( \ell_{\text{max}} = 10 \) for the perpendicular configuration since the curves are converging near each \( \ell_{\text{max}} \). This convergence becomes more vivid for a certain region of \( \omega \) for each configuration.

![Graph](image)

Figure 5.10: The radial component of the time-averaged force between two identical spheres with \( \epsilon_i \), plotted as a function of frequency \( \omega/\omega_p \), where \( \omega_p \) is the plasma frequency, for different \( \ell_{\text{max}} \). It is assumed that \( \epsilon_i \) follows the Drude model with \( \omega_p \tau = 20 \) and that the host material has \( \epsilon_h = 1 \). The gap spacing between two spheres is \( \delta = 0.01 \) mm. The applied electric field is \( E_0 = 25.2 \) V/mm and \( a = 3.15 \) mm. The electric field is parallel to the line between two spheres in (a) and perpendicular to that line in (b).

### 5.4 Discussion

The present work permits calculation of electrical forces in electrorheological materials in a concise closed form, which permits inclusion of all multipoles and all
many-body forces in a simple way. In our approach, the forces do not need to be calculated as numerical derivatives; instead, we give explicit analytical expressions for these derivatives, in terms of a characteristic pole spectrum which describes the geometry of the material. The explicit form for the derivatives is somewhat reminiscent of the Hellmann-Feynman description of quantum-mechanical forces in electronic structure theory, but differs from it in important ways.

One striking feature of the present formalism is that it allows for the calculation of frequency-dependent forces in a simple closed form. Although such forces have been discussed in previous work [42, 172, 171, 114], the present approach is relatively simple and more general, and places both zero and finite frequency forces within the same formalism. In our numerical work, we find that these forces can even change sign as a function of frequency. Such frequency-dependence is, of course, also present in the long-range (dipole-dipole) limit treated by others in previous work, but it is even more apparent in this study.

Although in the present chapter the calculations have been carried out explicitly for two-body interactions, they can readily be extended to three-body (or multibody) forces, too. The general equation (5.30) can be used to find the force on a sphere, no matter how many particles are contained in the suspension. Indeed, such multibody forces are very likely to play important roles in dense suspensions, where they could possibly lead to “bond-angle-dependent” forces analogous to those well-known in liquid and solid semiconductors. Likewise, the calculations could be readily extended to other types of shape (e. g., hollow spherical shells), provided that the requisite pole spectra and matrix elements can be calculated. In principle, one could even deal also with nonspherical shapes, though the calculation of the pole spectrum in this
case would likely be more difficult. Also, although we have restricted our calculations in this chapter to forces directed along a line between two particles, it would be straightforward to calculate other components of the force, which could give rise to a torque with respect to that line. Finally, the present formalism can be immediately extended to the important case of magnetorheological fluids. For such fluids, Eq. (5.30) for the force would continue to be valid, provided that $\epsilon_i$ and $\epsilon_h$ are replaced by $\mu_i$ and $\mu_h$.

It is useful to add some comments about the details of our calculations for some real materials studied in recent experiments. Our calculated frequency-dependence of the forces in the case of SrTiO$_3$ spheres may appear to disagree with that obtained in Refs. [185, 184] at close spacing. There are a number of plausible explanations for this discrepancy. One likely possibility is that the host fluid does not have a homogeneous conductivity when two highly polarizable spheres are placed in it in close proximity. Instead, there could well be nonlinear screening effects of the Debye-Hückel type [46, 45], which would mean that the picture of a two-component composite is simply not appropriate in this regime. In support of this hypothesis, we note that the reported experimental forces are still frequency-dependent at high frequencies, while the complex dielectric functions of both host and sphere should be nearly frequency-independent in this regime, leading to a frequency-independent force in this range. A second, and perhaps the most plausible, explanation is that there are not enough data provided in Refs. [185, 184] in the low frequency region to be compared with our numerical results. Most of our calculations were carried out in the very low frequencies, less than 100 Hz. The experimental plots include at most one or two points at discrete frequencies in this range.
The present method could readily be combined with standard molecular dynamics approaches to compute dynamical properties of electrorheological (or magnetorheological) fluids. Specifically, one could carry out molecular dynamics (MD) calculations, following the approach of several authors [96, 95, 22, 94, 72, 173]. In such approaches, the force on a given sphere is typically expressed as the sum of a hard-sphere repulsion, a viscous force, and an electrostatic force. The first two of these forces would be the same as in the previous MD studies, but the third would be calculated using the present method, rather than the dipole-dipole force generally used in other MD studies. It would be of great interest to see how such quantities as viscous relaxation time would be affected by using our forces in these calculations. In addition to such calculations, it would be of interest to study minimum-energy configurations of dielectric suspensions in an applied electric field, based on the forces calculated using the methods outlined here. Many such studies can already be found in the literature (see, e. g., Ref. [41] or [33]). Corrections arising from nondipolar interactions will clearly be quite important here also.
CHAPTER 6

PHOTONIC BAND STRUCTURES OF METAL INVERSE OPALS

6.1 Introduction

The photonic band structures of composite materials have been studied extensively. Such band structures are defined by the relation between frequency $\omega$ and Bloch vector $\mathbf{k}$ in media in which the dielectric constant is a periodic function of position. A major reason for such interest is the possibility of producing photonic band gaps, i.e., frequency regions, extending through all $\mathbf{k}$-space, where electromagnetic waves cannot propagate through the medium. Such media have many potentially valuable applications, including possible use as filters and in films with rejection-wavelength tuning [190]. In systems with a complete photonic band gap, the spontaneous emission of atoms with level splitting within the gap can be strongly suppressed [25].

The photonic band structure of a range of materials has been studied using a plane wave expansion method. Typically, the method converges easily when the dielectric function is everywhere real, but more slowly, or not at all, when the dielectric function has a negative real part, as occurs when one component is metallic. For example, McGurn et al. [122] used this method to calculate the photonic band structure of a
square lattice of metal cylinders in 2D and of an fcc lattice of metal spheres embedded in vacuum in 3D. They found that that method converged well when the filling fraction \( f \) (i.e., volume fraction of metal spheres or cylinders) satisfied \( f \leq 0.1\% \).

Kuzmiak et al. [105] used the same method to calculate the photonic band structures for 2D metal cylinders in a square or triangular lattice in vacuum. For low \( f \) and \( \omega > \omega_p \), the calculated photonic band structures are just slightly perturbed versions of the dispersion curves for electromagnetic waves in vacuum. However, for \( \omega < \omega_p \) and \( \mathbf{H} \)-polarized waves (magnetic field \( \mathbf{H} \) parallel to the cylinders), they obtained many nearly flat bands for \( \omega < \omega_p \); these bands were found to converge very slowly with increasing numbers of plane waves. They later extended this work to systems with dissipation [104]. To describe dispersive and absorptive materials, they used a complex, position-dependent form of dielectric function. They also introduced a standard linearization technique to solve the resulting nonlinear eigenvalue problem.

Zabel et al. [195] extended the plane wave method to treat periodic composites with anisotropic dielectric functions. In particular, they studied the photonic band structures of a periodic array of anisotropic dielectric spheres embedded in air. They found that the anisotropy split degenerate bands, and narrowed or even closed the band gaps. Much further work on anisotropic photonic materials has been carried out since this paper [25].

Very recently, Gaillot et al. [57] have studied the photonic band structures of another type of structure, a so-called inverse opal structure. This structure is an fcc lattice of void spheres in a host of another material. Such a structure can be prepared, e.g., starting from an opal structure made of spheres of a convenient substance, infiltrating it with another material, then dissolving away the spheres. In
the work of Ref. [57], the photonic band structure of Si inverse opal was calculated 
as a function of the infiltrated volume fraction $f$ of air voids using three-dimension-
al finite difference time domain (3D FDTD) method. It was found that for certain 
values of $f$, a complete band gap opens up between the eighth and ninth bands.

In the present chapter, we study the photonic band structure of an inverse opal 
structure, such as that investigated in Ref. [57], but instead of dielectric materials 
such as Si, we consider metals as the infiltrated materials. Thus the material we 
study is also the inverse of the fcc array of metal spheres studied by McGurn et al. 
[122]. Such metallic inverse opal structures have recently become of great interest, 
because it has been found that Pb inverse opals exhibit superconductivity [3]. These 
workers have studied the response of these materials to an applied magnetic field, and 
have found a highly nonmonotonic fractional flux penetration into the Pb spheres as 
a function of the applied field.

Our primary method for studying the photonic band structure of metallic inverse 
opal is the plane wave expansion method described earlier. We find, as did McGurn 
et al. [122], that this method converges well for $\omega > \omega_p$ but much less so for $\omega < \omega_p$, 
where $\omega_p$ is the plasma frequency. For this reason, we also supplement our plane wave 
calculations with some calculations of the individual sphere modes. A somewhat sim-
ilar problem to ours has already been studied in 2D by Lee et al. [110]. These authors 
calculated the photonic band structure of a periodic square array of air cylinders 
embedded in a superconducting host, using a plane wave expansion method. They 
showed that there existed propagating bands of modes below the energy gap of the 
superconducting host when its $c$-axis was parallel to the direction of polarization. 
Our calculations obtains similar results for the metallic inverse opal structure.
The remainder of this chapter is organized as follows. In Sec. 6.2, we present the formalism for calculating the photonic band structures of metal inverse opals and for obtaining transverse magnetic and transverse electric (TM and TE) modes for a single spherical cavity. We also describe a simple tight-binding approach for $\omega < \omega_p$. In Sec. 6.3, we give the results of our calculations using a plane wave expansion for metallic inverse opals, of asymptotic calculations for a spherical cavity, and of the tight-binding method. Section 6.4 presents a summary and discussion.

6.2 Formalism

In this section, we present a summary of the equations determining the band structure of a photonic crystal containing a metallic component, with Drude dielectric function $\epsilon(\omega) = 1 - \omega_p^2/\omega^2$ and an insulating component of dielectric constant unity. We first give the equations based on a plane wave expansion of the electric field $\mathbf{E}$, then the corresponding equations based on expanding the magnetic induction $\mathbf{B}$ in plane waves. Next, we write down the equations for the TM and TE modes of a spherical cavity in a Drude metal. Finally, we present a tight-binding method for $\omega < \omega_p$.

6.2.1 Photonic Equations Based on the E Field

To obtain equations for the electric field $\mathbf{E}$, we start from the two homogeneous Maxwell equations

$$\nabla \times \mathbf{E} = \frac{i \omega}{c} \mathbf{B},$$  \hspace{1cm} (6.1)

$$\nabla \times \mathbf{B} = -\frac{i \omega}{c} \epsilon \mathbf{E}.$$  \hspace{1cm} (6.2)
Taking the curl of Eq. (6.1) and inserting Eq. (6.2) into this equation, we obtain

\[
\nabla \times (\nabla \times \mathbf{E}) = \frac{i \omega}{c} \nabla \times \mathbf{B} = \frac{\omega^2}{c^2} \epsilon \mathbf{E} \\
= \frac{\omega^2}{c^2} \left\{ \left(1 - \frac{\omega_p^2}{\omega^2}\right) \theta(\mathbf{x}) + 1 - \theta(\mathbf{x}) \right\} \mathbf{E} \\
= \frac{\omega^2}{c^2} \left(1 - \frac{\omega_p^2}{\omega^2} \theta(\mathbf{x})\right) \mathbf{E},
\]

(6.3)

where we have defined a step function \(\theta(\mathbf{x}) = 1\) inside a metal and \(\theta(\mathbf{x}) = 0\) elsewhere.

Since for a periodic lattice \(\mathbf{E}(\mathbf{x})\) is a Bloch function and \(\theta(\mathbf{x})\) is periodic, we can expand both in momentum space as

\[
\mathbf{E}(\mathbf{x}) = \sum_{\mathbf{K}'} \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) e^{i(\mathbf{k} + \mathbf{K}') \cdot \mathbf{x}},
\]

(6.4)

\[
\theta(\mathbf{x}) = \sum_{\mathbf{G}} \theta_{\mathbf{G}}(\mathbf{k}) e^{i \mathbf{G} \cdot \mathbf{x}},
\]

(6.5)

where \(\mathbf{K}'\) and \(\mathbf{G}\) are reciprocal lattice vectors. Inserting Eqs. (6.4) and (6.5) back into Eq. (6.3) and using the relations

\[
\nabla \times \mathbf{E} = \sum_{\mathbf{K}'} i(\mathbf{k} + \mathbf{K}') e^{i(\mathbf{k} + \mathbf{K}') \cdot \mathbf{x}} \times \mathbf{E}_{\mathbf{K}'}(\mathbf{k}),
\]

(6.6)

\[
\nabla \times \nabla \times \mathbf{E} = -\sum_{\mathbf{K}'} \left[ (\mathbf{k} + \mathbf{K}') \cdot \left( (\mathbf{k} + \mathbf{K}') \cdot \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) \right) - |\mathbf{k} + \mathbf{K}'|^2 \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) \right] \\
\times e^{i(\mathbf{k} + \mathbf{K}') \cdot \mathbf{x}},
\]

(6.7)

we get

\[
-\sum_{\mathbf{K}'} \left[ (\mathbf{k} + \mathbf{K}') \cdot \left( (\mathbf{k} + \mathbf{K}') \cdot \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) \right) - |\mathbf{k} + \mathbf{K}'|^2 \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) \right] e^{i(\mathbf{k} + \mathbf{K}') \cdot \mathbf{x}}
\]

\[
= \frac{\omega^2}{c^2} \left( \sum_{\mathbf{K}'} \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) e^{i(\mathbf{k} + \mathbf{K}') \cdot \mathbf{x}} - \frac{\omega_p^2}{\omega^2} \sum_{\mathbf{G}} \sum_{\mathbf{K}'} \mathbf{E}_{\mathbf{K}'}(\mathbf{k}) \theta_{\mathbf{G}}(\mathbf{k}) e^{i(\mathbf{G} + \mathbf{k} + \mathbf{K}') \cdot \mathbf{x}} \right).
\]

(6.8)
Multiplying both sides of Eq. (6.8) by \( \exp[-i(k + K) \cdot x] \), integrating over volume \( v_c \) of a primitive unit cell, and simplifying, we obtain

\[
- \left[ (k + K) \{ (k + K) \cdot E_K(k) \} - |k + K|^2 E_K(k) \right] = \frac{\omega^2}{c^2} \left( E_K(k) - \frac{\omega^2}{\omega^2} \sum_{K'} E_{K'}(k) \theta_{K'-K} \right),
\]

(6.9)

where

\[
\theta_{K'-K} = \frac{1}{v_c} \int_{v_c} \theta(x) e^{-i(K - K') \cdot x} dx
\]

= \delta_{K'-K,0} - (1 - \delta_{K'-K,0}) \frac{1}{v_c} \int_v e^{-i(K - K') \cdot x} dx,
\]

(6.10)

here \( v \) is the volume of a void sphere centered at the origin. The second term of Eq. (6.10) is readily evaluated using

\[
\int_v e^{-iK \cdot r} dr = \frac{4\pi}{K^3} \left[ \sin(KR) - KR \cos(KR) \right],
\]

(6.11)

where \( R \) is the radius of the void sphere centered at the origin and \( K = |K| \). In dimensionless form, Eq. (6.9) can be rewritten as

\[
-(k + K) \{ (k + K) \cdot E_K(k) \} + |k + K|^2 E_K(k) + \left( \frac{\omega p a}{2\pi c} \right)^2 \sum_{K'} E_{K'}(k) \theta_{K'-K}
\]

= \frac{\omega^2}{c^2} E_K(k),
\]

(6.12)

where \( \omega' = \omega a/(2\pi c) \) is an eigenvalue of Eq. (6.12), and \( k, K, \) and \( K' \) are all expressed in units of \( 2\pi/a \).

### 6.2.2 Photonic Equations Based on the B Field

The photonic band structure equations can also be expressed in terms of the \( B \) field rather than the \( E \) field. In this case, we combine Eq. (6.1) and Eq. (6.2) to obtain a single equation for \( B \):

\[
\nabla \times \left[ \left\{ \frac{1}{1 - \frac{\omega^2}{\omega^2}} \theta(x) + 1 - \theta(x) \right\} \nabla \times B \right] = \frac{\omega^2}{c^2} B,
\]

(6.13)
where we have used \(1/\varepsilon(x, \omega) = \theta(x)/(1 - \omega_p^2/\omega^2) + 1 - \theta(x)\). Multiplying by \(\omega^2 - \omega_p^2\) and simplifying, we obtain

\[
\omega_p^2 \nabla \theta(x) \times (\nabla \times \mathbf{B}) + [\omega^2 - \omega_p^2(1 - \theta(x))] \nabla \times (\nabla \times \mathbf{B}) = \frac{\omega^2}{c^2} (\omega^2 - \omega_p^2) \mathbf{B}.
\]  

(6.14)

Introducing the Fourier-transform of \(\mathbf{B}(x) = \sum_{\mathbf{k}} \mathbf{b}_{\mathbf{k}}(k) \exp[i(\mathbf{k} + \mathbf{k}') \cdot x]\), we obtain, after some simplification,

\[
-\omega_p^2 \sum_{\mathbf{k}'} \left\{ [(\mathbf{k} - \mathbf{k}') \cdot \mathbf{b}_{\mathbf{k}'}(k)(k + \mathbf{k}') - [(\mathbf{k} - \mathbf{k}') \cdot (k + \mathbf{k}')] \mathbf{b}_{\mathbf{k}'}(k) \right\} \theta_{\mathbf{k} - \mathbf{k}'} - (\omega^2 - \omega_p^2) \{ (\mathbf{k} + \mathbf{k}) \cdot \mathbf{b}_{\mathbf{k}}(k) - \mathbf{b}_{\mathbf{k}}(k)|k + \mathbf{k}|^2 \}
\]

\[
-\omega_p^2 \sum_{\mathbf{k}'} \theta_{\mathbf{k} - \mathbf{k}'} \{ (\mathbf{k} + \mathbf{k}') [((\mathbf{k} + \mathbf{k}') \cdot \mathbf{b}_{\mathbf{k}'}(k)) - \mathbf{b}_{\mathbf{k}'}(k)|k + \mathbf{k}'|^2 \}
\]

\[
= \frac{\omega^2}{c^2} (\omega^2 - \omega_p^2) \mathbf{b}_{\mathbf{k}}(k).
\]  

(6.15)

This last equation was obtained by multiplying both sides of the differential equation for \(\mathbf{B}(x)\) by \(\exp[-i(\mathbf{k} + \mathbf{k}') \cdot x]\), integrating over \(v_c\), and simplifying.

We can rearrange the various terms in Eq. (6.15) to obtain a matrix equation for the components of the magnetic field, using dimensionless wave vectors \(\mathbf{k}, \mathbf{K},\) and \(\mathbf{K}'\), and a dimensionless frequency \(\omega' = \omega a/(2\pi c)\). The final form of the equation is

\[
\mathbf{B}_\mathbf{k}(k) \omega' \mathbf{I} + \left\{ (\mathbf{k} + \mathbf{K}) [(\mathbf{k} + \mathbf{K}) \cdot \mathbf{B}_\mathbf{k}(k)] - \left\{ |\mathbf{k} + \mathbf{K}|^2 + \left( \frac{\omega_p \mu_r a}{2\pi \varepsilon} \right)^2 \right\} \mathbf{B}_\mathbf{k}(k) \right\} \omega'^2
\]

\[
+ \left( \frac{\omega_p \mu_r a}{2\pi \varepsilon} \right)^2 \sum_{\mathbf{k}'} \left\{ (\mathbf{k} + \mathbf{K}') [(\mathbf{k} - \mathbf{k}') \cdot \mathbf{B}_{\mathbf{k}'}(k)] + (\mathbf{k} + \mathbf{K}')(\mathbf{k} + \mathbf{K}') \cdot \mathbf{B}_{\mathbf{k}'}(k) \right\}
\]

\[- \left\{ [(\mathbf{k} - \mathbf{k}') \cdot \mathbf{b}_{\mathbf{k}'}(k)] + |k + \mathbf{k}'|^2 \right\} \theta_{\mathbf{k} - \mathbf{k}'}
\]

\[- \left\{ (\mathbf{k} + \mathbf{K}) [(\mathbf{k} + \mathbf{K}) \cdot \mathbf{b}_{\mathbf{k}'}(k)] - |k + \mathbf{K}'|^2 \mathbf{b}_{\mathbf{k}'}(k) \right\} = 0,
\]  

(6.16)

where the components of \(\mathbf{k}, \mathbf{K},\) and \(\mathbf{K}'\) are in units of \(2\pi/a\).

Equation (6.16) is a quartic equation in \(\omega'\) and can be simply rewritten symbolically as

\[
\omega'^4 \mathbf{I} + \omega'^2 \mathbf{P} + \mathbf{Q} = 0,
\]  

(6.17)
where $I$ is the identity matrix and $P$ and $Q$ are the coefficient matrices of $\omega^2$ and the constant term, respectively. To use the linearization technique of Ref. [104], we introduce the vector

$$B_K^*(k) = \begin{pmatrix} \omega^2 B_K(k) \\ B_K(k) \end{pmatrix}.$$ (6.18)

Then Eq. (6.17) can be represented in a matrix form as

$$\begin{pmatrix} -P & -Q \\ I & 0 \end{pmatrix} B_K^*(k) = \omega^2 B_K^*(k).$$ (6.19)

In this form, Eq. (6.19) corresponds to a *linear eigenvalue problem*.

If we include $N$ plane waves, the submatrices $-P$ and $-Q$ can be expressed as an $N \times N$ matrix, each of whose elements is a $3 \times 3$ submatrix. The off-diagonal submatrices of $-P$ are all zeroes, while the diagonal ones of $-P$ and $-Q$ are readily calculated.

### 6.2.3 Spherical Cavity

Next, we discuss the photonic band structure of an inverse opal from the point of view of single-cavity modes. We first discuss the transverse magnetic (TM) modes of a single spherical cavity in a metallic host, then the transverse electric (TE) modes.

**TM Modes**

To discuss the TM modes, we consider the differential equation for $B$ obtained from Maxwell’s equations. From Eq. (6.14), we have

$$[\omega^2 - \omega_p^2 (1 - \theta(x))] \nabla \times (\nabla \times B) = \frac{\omega^2}{c^2} (\omega^2 - \omega_p^2) B.$$ (6.20)

Thus, inside the spherical void, we have

$$\nabla \times (\nabla \times B) = \frac{\omega^2}{c^2} B,$$ (6.21)
while inside the metal,
\begin{align*}
\nabla \times (\nabla \times \mathbf{B}) &= \frac{\omega^2 - \omega_p^2}{c^2} \mathbf{B}.
\end{align*}

The nonvanishing components of the solutions for \( \mathbf{B} \) and \( \mathbf{E} \) for Eq. (6.21) are \[83\]
\begin{align*}
B_{\phi,\text{in}}(r, \theta) &= \frac{u_\ell(r)}{r} P^1_\ell(\cos \theta), \\
E_{r,\text{in}} &= -\frac{ic \ell(\ell + 1)}{\omega r} \frac{u_\ell(r)}{r} P(\cos \theta), \\
E_{\theta,\text{in}} &= -\frac{ic}{\omega r} \frac{\partial u_\ell(r)}{\partial r} P^1_\ell(\cos \theta), \\
u_\ell(r) &= r[A_{\ell} j_\ell(k r) + B_{\ell} n_\ell(k r)],
\end{align*}

where \( k = \omega/c, j_\ell \) and \( n_\ell \) are spherical Bessel functions, and the subscripts \( \phi, r, \) and \( \theta \) denote components of the corresponding fields in spherical coordinates.

Likewise, the solutions of Eq. (6.22) will be
\begin{align*}
B_{\phi,\text{out}}(r, \theta) &= \frac{v_\ell(r)}{r} P^1_\ell(\cos \theta), \\
E_{r,\text{out}} &= -\frac{ic \ell(\ell + 1)}{\omega r} \frac{v_\ell(r)}{r} P(\cos \theta), \\
E_{\theta,\text{out}} &= -\frac{ic}{\omega r} \frac{\partial v_\ell(r)}{\partial r} P^1_\ell(\cos \theta), \\
v_\ell(r) &= r[C_{\ell} j_\ell(k' r) + D_{\ell} n_\ell(k' r)],
\end{align*}

where \( k' = \sqrt{\omega^2 - \omega_p^2}/c. \)

The condition that normal \( D \) and tangential \( E \) should be continuous at \( R \) gives the two conditions
\begin{align*}
u_\ell(R) &= v_\ell(R) \quad (6.25)
\end{align*}

and
\begin{align*}
\left. \frac{\partial u_\ell(r)}{\partial r} \right|_{r=R} &= \frac{1}{\left(1 - \frac{\omega^2}{\omega_p^2}\right)} \left. \frac{\partial v_\ell(r)}{\partial r} \right|_{r=R} = \frac{k^2}{k'^2} \left. \frac{\partial v_\ell(r)}{\partial r} \right|_{r=R}, \quad (6.26)
\end{align*}
where \( R \) is the radius of the spherical cavity. Since the fields at the center of the void sphere must be finite, we also have

\[
u_\ell(r) = r j_\ell(kr),
\]

(6.27)

where we have normalized the solution so that the coefficient \( A_\ell = 1 \). From Eq. (6.25) we have

\[
j_\ell(kR) = C_\ell j_\ell(k'R) + D_\ell n_\ell(k'R),
\]

(6.28)

whereas, from Eq. (6.26), we get

\[
k^2 [j_\ell(kR) + kR j'_\ell(kR)]
\]

\[= k^2 [C_\ell j_\ell(k'R) + D_\ell n_\ell(k'R) + k'R \{C_\ell j'_\ell(k'R) + D_\ell n'_\ell(k'R)\}].
\]

(6.29)

The coefficients \( C_\ell \) and \( D_\ell \) can be decided from the boundary conditions.

The results for \( \omega < \omega_p \) can be obtained by making the substitution \( k' \rightarrow ik' \), with \( k' \) real. In this case, the radial component of Eq. (6.22) takes the form

\[
\frac{\partial^2 u_\ell}{\partial r^2} - \left[ \frac{1}{c^2} (\omega_p^2 - \omega^2) + \frac{\ell(\ell + 1)}{r^2} \right] u_\ell = 0,
\]

(6.30)

which is the modified Bessel equation. The solutions of Eq. (6.30) are the spherical modified Bessel functions \( i_\ell \) and \( k_\ell \) (note that this \( k_\ell \) is different from the wave vectors \( k \) and \( k' \)). In this case, Eq. (6.24) becomes

\[
v_\ell(r) = r [C_\ell i_\ell(k'r) + D_\ell k_\ell(k'r)],
\]

(6.31)

where \( k' = \sqrt{\omega_p^2 - \omega^2}/c \). In addition, Eqs. (6.26), (6.28), and (6.29) are transformed, respectively, into

\[
\frac{\partial u_\ell(r)}{\partial r} \bigg|_{r=R} = -\frac{k^2}{k'^2} \frac{\partial v_\ell(r)}{\partial r} \bigg|_{r=R},
\]

(6.32)
\[ j_\ell(kR) = C_\ell i_\ell(k'R) + D_\ell k_\ell(k'R), \]  

(6.33)

and

\[
k^2 [j_\ell(kR) + kR j'_\ell(kR)] = -k^2 [C_\ell i_\ell(k'R) + D_\ell k_\ell(k'R) + k'R \{C_\ell i'_\ell(k'R) + D_\ell k'_\ell(k'R)\}].
\]  

(6.34)

It is of interest to consider the specific case of a spherical cavity in an infinite medium. In this case, \( C_\ell = 0 \) because \( i_\ell(x) \) diverges at large \( x \). As a result, Eqs. (6.31), (6.33), and (6.34) become, respectively,

\[ v_\ell(r) = r D_\ell k_\ell(k'r), \]  

(6.35)

\[ j_\ell(kR) = D_\ell k_\ell(k'R), \]  

(6.36)

and

\[
k^2 [j_\ell(kR) + kR j'_\ell(kR)] = -k^2 D_\ell [k_\ell(k'R) + k'R k'_\ell(k'R)].
\]  

(6.37)

From Eq. (6.36) we have \( D_\ell = j_\ell(kR)/k_\ell(k'R) \), and hence Eq. (6.37) becomes

\[
k^2 [j_\ell(kR) + kR j'_\ell(kR)] = -k^2 \frac{j_\ell(kR)}{k_\ell(k'R)} [k_\ell(k'R) + k'R k'_\ell(k'R)].
\]  

(6.38)

We can readily obtain the asymptotic forms of the solutions when \( kR \ll 1 \) and \( k'R \ll 1 \). In this case \( j_\ell(kR) \) and \( k_\ell(k'R) \) have the asymptotic forms \( j_\ell(kR) \approx (kR)^\ell/(2\ell + 1)!! \) and \( k_\ell(k'R) \approx \sqrt{\pi} \Gamma(\ell + 1/2)2^{\ell-1}/(k'R)^{\ell+1} \). In this limit, Eq. (6.38), after some algebra, reduces to simply

\[ k^2(\ell + 1) = k^2 \ell. \]  

(6.39)

Since \( k' = \sqrt{\omega_p^2 - \omega^2}/c \), Eq. (6.39) is equivalent to

\[ \omega^2 = \frac{\ell + 1}{2\ell + 1} \omega_p^2. \]  

(6.40)

The largest value, \( \omega = \sqrt{2/3} \omega_p \), occurs at \( \ell = 1 \) and the limiting value for large \( \ell \) is \( \omega = \omega_p/\sqrt{2} \).
TE Modes

For the TE mode, inside the spherical void, we have

\[ \nabla \times (\nabla \times \mathbf{E}) = \frac{\omega^2}{c^2} \mathbf{E}, \quad (6.41) \]

whereas inside the metal, we have

\[ \nabla \times (\nabla \times \mathbf{E}) = \frac{\omega^2 - \omega_p^2}{c^2} \mathbf{E}. \quad (6.42) \]

We now use these equations to calculate \( E_\phi, B_r, \) and \( B_\theta \). From Eq. (6.41) we get

\[
E_{\phi,\text{in}}(r, \theta) = \frac{u_\ell(r)}{r} P^1_\ell(\cos \theta), \\
B_{r,\text{in}} = \frac{ic}{\omega r} \ell(\ell + 1) \frac{u_\ell(r)}{r} P_\ell(\cos \theta), \\
B_{\theta,\text{in}} = \frac{ic}{\omega r} \frac{\partial u_\ell(r)}{\partial r} P^1_\ell(\cos \theta), \\
u_\ell(r) = r [A_\ell j_\ell(kr) + B_\ell n_\ell(kr)], \quad (6.43)
\]

where \( k = \omega/c \). The solutions of Eq. (6.42) are

\[
E_{\phi,\text{out}}(r, \theta) = \frac{v_\ell(r)}{r} P^1_\ell(\cos \theta), \\
B_{r,\text{out}} = \frac{ic}{\omega r} \ell(\ell + 1) \frac{v_\ell(r)}{r} P_\ell(\cos \theta), \\
B_{\theta,\text{out}} = \frac{ic}{\omega r} \frac{\partial v_\ell(r)}{\partial r} P^1_\ell(\cos \theta), \\
v_\ell(r) = r [C_\ell j_\ell(k'r) + D_\ell n_\ell(k'r)], \quad (6.44)
\]

where \( k' = \sqrt{\omega^2 - \omega_p^2}/c \).

Since normal \( B \) and tangential \( H \) should be continuous on the boundaries, we obtain the conditions

\[ u_\ell(R) = v_\ell(R), \quad (6.45) \]
as in the TM case, and
\[ \frac{\partial u_\ell(r)}{\partial r} \bigg|_{r=R} = \frac{\partial v_\ell(r)}{\partial r} \bigg|_{r=R}. \] (6.46)
Since the fields must be finite at the center of the void sphere, we can choose
\[ u_\ell(r) = r j_\ell(kr), \] (6.47)
we also take the coefficient \( A_\ell = 1 \). From Eq. (6.45) we have
\[ j_\ell(kR) = C_\ell j_\ell(k'R) + D_\ell n_\ell(k'R), \] (6.48)
while, from Eq. (6.46), we get
\[ j_\ell(kR) + kRj'_\ell(kR) = C_\ell i_\ell(k'R) + D_\ell k_\ell(k'R) + k'R \{ C_\ell i'_\ell(k'R) + D_\ell k'_\ell(k'R) \}. \] (6.49)

The corresponding equation for \( \omega < \omega_p \) can again be obtained by the transformation \( k' \rightarrow ik' \). The TE modes for \( \omega < \omega_p \) using the modified spherical Bessel functions \( i_\ell(x) \) and \( k_\ell(x) \) satisfy Eqs. (6.30), (6.31), (6.46), and (6.33). The only changes are in Eq. (6.34), which becomes
\[ j_\ell(kR) + kRj'_\ell(kR) = C_\ell i_\ell(k'R) + D_\ell k_\ell(k'R) + k'R \{ C_\ell i'_\ell(k'R) + D_\ell k'_\ell(k'R) \}. \] (6.50)

In an infinite medium, these conditions become, from Eq. (6.36),
\[ j_\ell(kR) + kRj'_\ell(kR) = \frac{j_\ell(kR)}{k_\ell(k'R)} \{ k_\ell(k'R) + k'Rk'_\ell(k'R) \}. \] (6.51)

If we consider the asymptotic forms of the solutions when \( kR \ll 1 \) and \( k'R \ll 1 \) as we did in the TM modes, Eq. (6.51) ends up with
\[ \ell + 1 = -\ell, \] (6.52)
which gives \( \ell = -1/2 \). Therefore the eigenvalues don’t exist for TE modes in this limit.
6.2.4 Tight-Binding Approach to Modes for $\omega < \omega_p$

Since the plane wave basis seems to converge poorly for modes with frequencies $\omega < \omega_p$, it seems worthwhile to try a different approach. This is the tight-binding method, which is very useful in treating narrow bands in conventional solids. In what follows, we try to suggest a tight-binding approach for the lowest set of TM modes in an fcc lattice.

Our work already gives the equations determining isolated TM modes. The lowest set corresponds to $\ell = 1$, and there should be three of these. The electric fields of these three modes should be orthogonal, i.e.,

$$\int E_\lambda^*(x) \cdot E_\mu(x) \, dx = \delta_{\lambda,\mu}, \quad (6.53)$$

where $E_\lambda$ and $E_\mu$ are the normalized electric fields for modes $\mu$ and $\lambda$. If the frequencies of the modes are different, they can be shown to be orthogonal, and if they are not different, then one can choose an orthonormal set. Let’s denote these three modes $E_1, E_2,$ and $E_3$. We need to calculate matrix elements of the form

$$M_{\alpha,\beta}(R) = \int E_\alpha^*(x) \cdot O E_\beta(x - R) \, dx, \quad (6.54)$$

corresponding to two modes associated with different cavities centered at the origin and at $R$. Here, $O$ is the “Hamiltonian” of the system and satisfies

$$O E_\mu(x) = \nabla \times (\nabla \times E_\mu(x)) + \frac{\omega_p^2 \theta(x)}{c^2} E_\mu(x). \quad (6.55)$$

To calculate the Bloch states corresponding to the three $\ell = 1$ single-cavity modes, we introduce normalized Bloch states

$$E_{k,\lambda}(x) = \sum_R e^{ik \cdot R} E_\lambda(x - R), \quad (6.56)$$
where $k$ is a Bloch vector, and $R$ are the Bravais lattice vectors. We also introduce the elements of the “Hamiltonian” matrix

$$M_{\lambda,\mu}(k) = \sum_{R} e^{i k R} M_{\lambda,\mu}(R). \quad (6.57)$$

Finally, we make the usual tight-binding assumption that the “atomic” states corresponding to different cavities are orthogonal:

$$\int E^*_\lambda(x - R) \cdot E_{\mu}(x - R') dx = \delta_{\lambda,\mu} \delta_{R,R'}. \quad (6.58)$$

Although we have written one of the electric fields as a complex conjugate, this operation is not necessary, since the single-cavity fields are real for states below the plasma frequency. This orthogonality of states on different cavities is reasonable, since the fields fall off exponentially with separation.

Therefore, we obtain the frequencies by diagonalizing a $3 \times 3$ matrix as follows:

$$\text{det} \left[ M_{\lambda,\mu}(k) - \left( \frac{\omega^2(k)}{c^2} - \frac{\omega^2_{at}}{c^2} \right) \delta_{\lambda,\mu} \right] = 0, \quad (6.59)$$

where $\omega_{at}$ is the eigenvalue of a single-cavity. The solutions to these equations give the three $p$-bands for a periodic lattice of cavities in a metallic host.

We briefly comment on the connection between this approach and that used by earlier workers [23, 141]. In this work, the authors treat wave propagation along a chain of metallic nanoparticles. They use the tight-binding approximation, as we do, but in the quasistatic approximation in which one assumes that $\nabla \times E = 0$. This approximation is reasonable when both the particle radii and the interparticle separations are small compared to a wavelength, but is not accurate in other circumstances. Furthermore, even in the small-particle and small-separation regime, this approximation still fails to account for the radiation which occurs at certain wave numbers and
frequencies. The present approach would generalize this tight-binding method to (a) three dimensions instead of one; (b) pore modes instead of small particle modes; and most importantly (c) larger pores and larger interparticle separations.

Next, we discuss the numerical evaluation of the required matrix elements, Eq. (6.54). The relevant electric fields are given in this chapter, but in spherical coordinates. It should not be difficult to convert these into Cartesian coordinates. The operator $O$ is just a little trickier. We first note that $O = O_0 + O'$, where $O_0$ is the single-cavity operator which includes only that part of the $\theta$ function referring to $R$th site. But

$$O_0 E_\beta(x - R') = \frac{\omega^2}{c^2} E_\beta(x - R'),$$

(6.60)

since $E_\beta$ is an eigenstate of $O_0$ with eigenvalue $\omega^2/c^2$.

Since we are assuming that the overlap integral between electric field states centered on different sites vanishes, this term does not contribute to the matrix element $M_{\alpha,\beta}$, which is therefore just given by

$$M_{\alpha,\beta}(R) = \int E_\alpha(x) \cdot O' E_\beta(x - R) dx.$$  

(6.61)

We can also write

$$O' = \frac{\omega_p^2}{c^2} \sum_{R'} \theta_{R'}(x),$$

(6.62)

where

$$\theta_{R'} = \theta(x - R')$$

(6.63)

is a step function which is unity inside the cavity centered at $R'$ and is zero otherwise.

A reasonable approximation to Eq. (6.62) might be to include just $R' = 0$. In this case, we finally will get

$$M_{\alpha,\beta}(R) \sim \frac{\omega_p^2}{c^2} \int E_\alpha(x) \cdot E_\beta(x - R) dx,$$

(6.64)
where the integral runs just over the cavity centered at the origin. As a further approximation, we can just replace $E_{\beta}(x - R)$ by the value of this function at the origin, i.e., $E_{\beta}(-R)$. Then this field comes out of the integral and we just have

$$M_{\alpha,\beta}(R) \sim \frac{\omega^2}{c^2} E_{\beta}(-R) \cdot \int E_{\alpha}(x) d\mathbf{x}, \quad (6.65)$$

where once again the integral runs over the cavity centered at the origin.

Next, we attempt to calculate the relevant quantities needed to solve for this matrix element. In order to use the tight-binding approach we will need to normalize the individual eigenstates $E_{\alpha}$. Therefore, we will begin by obtaining this normalization. For $\ell = 1$, the $u_{\ell}(r)$’s are $r$ times spherical Bessel functions. We write this field as

$$E_{r,\text{in}} = + \frac{2C_1}{kr} j_1(kr) \cos \theta, \quad (6.66)$$

where we have used the relation $P_1(\cos \theta) = \cos \theta$ and introduced the normalization constant $C_1$, which will be determined below. Similarly,

$$E_{\theta,\text{in}} = - \frac{C_1}{kr} \frac{\partial [rj_1(kr)]}{\partial r} \sin \theta, \quad (6.67)$$

where we use $P_1^1(\cos \theta) = -\sin \theta$. For $r > R$, we have

$$E_{r,\text{out}} = + \frac{2D_1C_1}{k'r} k_1(k'r) \cos \theta \quad (6.68)$$

and

$$E_{\theta,\text{out}} = - \frac{D_1C_1}{k'r} \frac{\partial [rk_1(k'r)]}{\partial r} \sin \theta. \quad (6.69)$$

We will need the integrals of the Cartesian components of the field over the volume of the sphere centered at the origin. Let us assume we are considering the $z$ mode, i.e., the one for which $\theta$ refers to the angle from the $z$ axis. Then the symmetry of
the problem shows that only the z component of the electric field will have a nonzero integral. Also, we have that

\[ E_{z,\text{in}}(r, \theta) = E_{r,\text{in}} \cos \theta - E_{\theta,\text{in}} \sin \theta. \]  

(6.70)

Thus, after a little algebra, we find that the integral of this field over the volume of the cavity is

\[ \int E_{z,\text{in}}(r, \theta) \, dr = \frac{8\pi C_1}{3k} R^2 j_1(kR). \]  

(6.71)

Next, we work out the coefficient \( D_1 \). It is determined by the boundary conditions at \( r = R \). These conditions are that \( D_r \) and \( E_\theta \) should be continuous at \( r = R \). These two conditions determine not only the value of \( D_1 \) but also the allowed frequency. After a bit of algebra, we find that

\[ D_1 = \frac{\omega^2 j_1(kR)}{c^2 k k'} \frac{k}{k' k_1(k'R)} \]  

(6.72)

The allowed value of \( \omega \) is given by Eq. (6.38).

Finally, we need the normalization constant \( C_1 \). We choose this so that the integral of the square of the electric field for a single-cavity mode should be normalized to unity. This condition may be written

\[ \frac{2C_1^2}{k^2} \frac{4\pi}{3} \int_0^R \left[ 2j_1^2(kr) + \left( \frac{\partial[r j_1(kr)]}{\partial r} \right)^2 \right] \, dr + \frac{2C_1^2 D_1^2}{k^2} \frac{4\pi}{3} \int_R^\infty \left[ 2k_1^2(k'r) + \left( \frac{\partial[r k_1(k'r)]}{\partial r} \right)^2 \right] \, dr = 1. \]  

(6.73)

If we write

\[ \int_0^{kR} \left[ 2j_1^2(x) + \left( \frac{d[x j_1(x)]}{dx} \right)^2 \right] \, dx = F_1(kR) \]  

(6.74)

and

\[ \int_{k'R}^{\infty} \left[ 2k_1^2(x) + \left( \frac{d[x k_1(x)]}{dx} \right)^2 \right] \, dx = F_2(k'R), \]  

(6.75)
then we can express the normalization condition as

\[
\frac{8\pi C_1^2}{3} \left[ \frac{F_1(kR)}{k^3} + D_1^2 \frac{F_2(k'R)}{k'^3} \right] = 1. \tag{6.76}
\]

Therefore, we can now write out an explicit expression for the matrix element \( M_{\alpha,\beta}(\mathbf{R}) \) given in Eq. (6.65). For the \( \alpha \)th mode, the integral of \( \mathbf{E}_\alpha \) over the volume of a cavity is a vector in the \( \alpha \)th direction. To evaluate Eq. (6.65), we need the component of the \( \alpha \)th mode in the \( \beta \)th direction at a position \( \mathbf{R} \). Let us first consider the \( z \) mode (\( \alpha = z \)). We can use Eqs. (6.68) and (6.69) to rewrite this field in Cartesian coordinates with the additional equations

\[
\begin{align*}
\cos \theta &= \frac{z}{r}, \\
\sin \theta &= \frac{\sqrt{x^2 + y^2}}{r}, \\
\hat{r} &= \frac{x\hat{x} + y\hat{y} + z\hat{z}}{r}, \\
\hat{\theta} &= \frac{xz\hat{x} + yz\hat{y}}{r \sqrt{x^2 + y^2}} - \frac{\sqrt{x^2 + y^2}}{r} \hat{z}.
\end{align*}
\tag{6.77}
\]

We just plug these expressions back into Eqs. (6.68) and (6.69) to get the Cartesian components of the field for a mode parallel to the \( z \) axis. For the mode parallel to the \( x \) axis, we just permute the coordinates cyclically: \( z \to x, \ x \to y, \) and \( y \to z \). Similarly, for the \( y \) modes, we make the permutation \( (x, y, z) \to (z, x, y) \).

Using these results, we should be able to compute all the elements in the tight-binding matrix and hence obtain the band structure for the photonic \( p \)-bands in the tight-binding approximation.
6.3 Numerical Results

Throughout this chapter we assume a lattice constant $a = 500\sqrt{2}$ nm and a void sphere radius $R = 150$ nm. Since the volume of the primitive unit cell is $v_c = a^3/4$, this corresponds to a void volume fraction $f = 0.160$.

Our band structures are expressed in terms of the standard notation for $k$ values at symmetry points in the Brillouin zone. These are $\Gamma = (0,0,0)$, $X = (2\pi/a)(0,0,1)$, $U = (2\pi/a)(1/4,1/4,1)$, $L = (2\pi/a)(1/2,1/2,1/2)$, $W = (2\pi/a)(1/2,0,1)$, and $K = (2\pi/a)(3/4,0,3/4)$.

The metallic dielectric function we assume for the inverse opals is the usual Drude form,

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2}, \quad (6.78)$$

where $\omega_p$ is the plasma frequency of the conduction electrons. $\epsilon(\omega) < 0$ when $\omega < \omega_p$, while $\epsilon(\omega) > 0$ when $\omega > \omega_p$. Our calculations are thus carried out assuming that the Drude relaxation time $\tau \to \infty$. For a metal in its normal state, $\omega_p^2 = 4\pi ne^2/m$, where $n$ is the conduction electron density and $m$ is the electron mass. Note that with this choice of dielectric function, the entire band structure can be expressed in scaled form. That is, the scaled frequency $\omega a/c$ is a function only of the scaled wave vector $ka$, and the band structures are parameterized by the two constants $\omega_p a/c$ and $f$. In our calculations, we include all plane waves corresponding to the set of reciprocal lattice vectors $\mathbf{K}$ satisfying the condition

$$0 \leq |\mathbf{K}| \leq K_c, \quad (6.79)$$
where $K_c$ is a given cutoff value. In the calculations described below, we have used $K_c a/(2\pi) = 7, 8, \text{ and } 9$. These values correspond to including 339, 537, and 749 plane waves, respectively, for the fcc lattice.

In Fig. 6.1 we plot the photonic band structure for a metallic inverse opal in the frequency range $\omega \geq \omega_p$, where we have used $\omega_p a/(2\pi c) = 7.694$ and included 537 plane waves. Partial photonic band gaps can be seen to occur in the $L$-$\Gamma$-$X$ region between the first and third bands and in the $U$-$X$-$W$-$K$ region between the ninth and tenth bands; however, there is no full photonic band gap.

![Figure 6.1: Photonic band structures for a metallic inverse opal with void radius $R = 150 \text{ nm}$ and lattice constant $a = 500\sqrt{2} \text{ nm}$ in the frequency range $\omega_p \leq \omega < 16\pi c/a$, where we use $\omega_p a/(2\pi c) = 7.694$. The number of plane waves used in this calculation is 537 and we expand the $E$ field of the modes.](image)

In Fig. 6.2, we show the corresponding photonic band structure for $\omega < \omega_p$, once again including 537 plane waves, particularly in the lower half of this frequency range. When this number of plane waves is included, the bands in this frequency range are
very flat, except near $\omega a/(2\pi c) = 1.5$ and at frequencies between 2.0 and 2.5. Because the bands are so flat, there are many gaps in which there appear to be no allowed photonic states. Furthermore, most of the bands are multiply degenerate.

Figure 6.2: Same as Fig. 6.1, except $\omega a/(2\pi c) \leq 3$. These bands correspond to $\omega < \omega_p$.

We have also recalculated the results of Figs. 6.1 and 6.2 using different numbers of plane waves. For $\omega \geq \omega_p$, the results are found not to depend on the number of plane waves used; increasing the number of plane waves just introduces additional bands at frequencies higher than the ones already calculated. However, when $\omega < \omega_p$, the calculated band structure does depend on the number of plane waves included, suggesting that the results shown have not fully converged. As an illustration of this behavior, we show in Fig. 6.3 the lowest few ($\leq 10$) bands for $\omega < \omega_p$, as calculated including 339, 537, and 749 plane waves. The bands are clearly seen to depend strongly on the number of plane waves included. As the number of plane waves
increases, the lowest band is taken up by the greatest number of plane waves, the next lowest band by the second greatest number of plane waves, and so on. We have also seen this trend when we have calculated the band structure using other values of $\omega_p$. These results illustrate the difficulty of obtaining fully converged results in a plane wave basis when $\omega < \omega_p$.

Figure 6.3: Same as Fig. 6.2, except that only a few ($\leq 10$) of the lowest eigenvalues are plotted for three different numbers of plane waves used: 339, 537, and 749, at frequencies such that $\omega < \omega_p$.

We have carried out additional calculations using other values of $\omega_p$, namely $\omega_p a/(2\pi c) = 1/(2\pi)$ and $\omega_p a/(2\pi c) = 1$. Fig. 6.4 shows the photonic band structures for $\omega_p a/(2\pi c) = 1/(2\pi)$ when $\omega > \omega_p$. The number of plane waves used here is 537 and our calculations are carried out in terms of the $\mathbf{E}$ field. Once again, the number of plane waves does not affect the band structure, just as in Fig. 6.1, at least
for the number of plane waves we consider here. When we calculate the band structure in terms of the $\mathbf{B}$ rather than $\mathbf{E}$ field, the results were unchanged, as they must be.

![Figure 6.4: Same as Fig. 6.1, except that $\omega_p < \omega < 6\pi c/a$ and $\omega_p a/c = 1$.](image)

For $\omega < \omega_p$, some representative examples of our numerical results, obtained using the $\mathbf{E}$ field, are shown in Fig. 6.5, while some others using the $\mathbf{B}$ field are shown in Fig. 6.6. In both cases, we include 537 plane waves. For the $\mathbf{E}$ field results, the bands appear to accumulate near $\omega = \omega_p$, while the results for the $\mathbf{B}$ field seem to show this type of accumulation both near $\omega = \omega_p$ and near $\omega = 0$. Most strikingly, in both cases, the bands are generally very flat, except for the $\mathbf{B}$ field results in the region between $\omega a/(2\pi c) = 0.12$ and 0.13, where some dispersion is evident.

In Figs. 6.7 and 6.8, we show the corresponding numerical results for $\omega_p a/(2\pi c) = 1$, for the two frequency regimes $\omega \geq \omega_p$ and $\omega < \omega_p$. The bands above $\omega_p$ are quite similar to those of $\omega_p a/c = 1$, except that many bands are highly degenerate in Fig.
Figure 6.5: Same as Fig. 6.4, except that $\omega a/(2\pi c) < 0.05$.

Figure 6.6: Same as Fig. 6.5, except that $0.2\pi c/a < \omega < \omega_p$ and we expand the $\mathbf{B}$
field of the modes.

6.7. Fig. 6.8 is also quite similar to Fig. 6.5 except that the bands seem to have more
degeneracies in Fig. 6.8.
Figure 6.7: Same as Fig. 6.4, except $\omega_p a/(2\pi c) = 1$.

Figure 6.8: Same as Fig. 6.7, except $\omega a/(2\pi c) < 0.5$.

Since we are considering void spheres in inverse opals, it is of interest to consider electromagnetic wave modes in a single cavity, which could be considered a single
“atom” of the void lattice. We show only results for $\omega < \omega_p$, since these are the results most relevant to possible narrow-band photonic states in the inverse opal structure. Our results for $\omega < \omega_p$ for an isolated spherical cavity in an infinite medium and when $kR \ll 1$ and $k'R \ll 1$ are given in Table 6.1. The latter condition is reasonable for our system parameters $a = 500\sqrt{2}$ nm, $R = 150$ nm, and $\omega_p a/c = 1$ according to

$$kR = \frac{\omega}{c} < \frac{\omega_p}{c} R = \frac{\omega_p a}{c} R = \frac{3}{10\sqrt{2}} = 0.2121,$$

$$k'R = \sqrt{\frac{\omega_p^2 - \omega^2}{c}} R = \sqrt{\left(\frac{\omega_p R}{c}\right)^2 - \left(\frac{\omega R}{c}\right)^2} = \sqrt{\left(\frac{\omega_p a}{c} R\right)^2 - (kR)^2}$$

$$= \sqrt{\left(\frac{3}{10\sqrt{2}}\right)^2 - (kR)^2} = \sqrt{0.045 - (kR)^2} < \sqrt{0.045} = 0.2121. \quad (6.80)$$

The (modified) spherical Bessel functions in Eq. (6.38) are extremely close to the $\omega$ axis for $\ell > 5$, so that it’s hard to get eigenfrequencies for $\ell > 5$ in the isolated spherical cavity. However the eigenfrequencies continue to exist for $\ell > 5$ when $kR \ll 1$ and $k'R \ll 1$.

<table>
<thead>
<tr>
<th>$\ell$</th>
<th>Infinite medium</th>
<th>$kR \ll 1$, $k'R \ll 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\ell = 1$</td>
<td>0.1296</td>
<td>0.1299</td>
</tr>
<tr>
<td>$\ell = 2$</td>
<td>0.1232</td>
<td>0.1233</td>
</tr>
<tr>
<td>$\ell = 3$</td>
<td>0.1203</td>
<td>0.1203</td>
</tr>
<tr>
<td>$\ell = 4$</td>
<td>0.1186</td>
<td>0.1186</td>
</tr>
<tr>
<td>$\ell = 5$</td>
<td>0.1178</td>
<td>0.1175</td>
</tr>
</tbody>
</table>

Table 6.1: TM mode frequencies $\omega' = \omega a/(2\pi c)$, where $\omega < \omega_p$ and $\omega_p a/c = 1$, calculated for an isolated spherical cavity and those when both $kR \ll 1$ and $k'R \ll 1$. The spherical Bessel functions are extremely close to the $\omega'$ axis for $\ell > 5$, so that it’s hard to get eigenfrequencies for $\ell > 5$ in the isolated spherical cavity. However this doesn’t happen when $kR \ll 1$ and $k'R \ll 1$. 

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The solutions for Eq. (6.51) don’t exist for \( \omega < \omega_p \) with \( \omega_p a/c = 1 \). This fact is consistent with that the eigenvalues for \( \omega < \omega_p \) don’t exist for TE modes when \( kR \ll 1 \) and \( k'R \ll 1 \).

For fcc, we include only the 12 nearest neighbors of the origin. Thus \( R = (a/2)(\pm 1, \pm 1, 0), (a/2)(\pm 1, \mp 1, 0), (a/2)(\pm 1, 0, \pm 1), (a/2)(0, \pm 1, \pm 1) \), and \( (a/2)(0, \pm 1, \mp 1) \). Assuming \( \omega_p a/c = 1 \) and using \( \omega_{at}(2\pi c) = 0.1296 \) for \( \ell = 1 \) in an infinite medium, we get the tight-binding results in Fig. 6.9, which shows three separate bands in the \( X-U-L \) region and \( X-W-K \) region as expected for the \( p \)-bands. The bandwidth is relatively small as \( M_{\alpha\beta}(R)a^2 \sim 0.001 \), which proves the general relation between the bandwidth and the overlap integral [6]. All three bands are degenerate at \( k = 0, \Gamma \) point. In addition, there is a double degeneracy when \( k \) is directed along either a cube axis (\( \Gamma-X \)) or a cube diagonal (\( \Gamma-L \)). A band gap occurs at the \( U \) point and the bands cross at the \( W \) point.

Figs. 6.10 and 6.11 show the tight-binding results plotted together with the plane wave expansions for the \( E \) field and for the \( B \) field, respectively. The \( p \)-bands are a little closer to the plane wave expansion for the \( B \) field. The plane wave expansion for the \( B \) field seems to be more appropriate to the problem in this range of eigenfrequencies.

### 6.4 Discussion

In this chapter we have calculated the photonic band structures of metal inverse opals above and below \( \omega_p \) using a plane wave expansion method based on either the \( E \) field or the \( B \) field of the photonic modes. We find that the band structures above \( \omega_p \) vary smoothly with \( k \) and do not depend on the number of plane waves
Figure 6.9: Tight-binding results for $\omega < \omega_p$ with $a = 500\sqrt{2}$ nm, $R = 150$ nm, and $\omega_p a/c = 1$, using $\omega_{\text{mt}} a/(2\pi c) = 0.1296$ for $\ell = 1$ in an infinite medium. The horizontal dotted line represents the atomic level.

Figure 6.10: Tight-binding results for $\omega < \omega_p$ plotted together with those from the plane wave expansion for the $E$ field. The number of plane waves used in the plane wave expansion is 537.
Figure 6.11: Same as Fig. 6.10, except the plane wave expansion for the B field.

included in the expansion, nor on the type of field used in the expansion. However, the bands for modes below \( \omega_p \) do depend on both, and are very flat. As a possible point of comparison, we have also calculated the TM and TE modes for a single spherical cavity using the asymptotic forms of spherical and modified spherical Bessel functions. These modes may account for some of the flat bands below \( \omega_p \) in metallic inverse opals. However there are no TE modes in the asymptotic limit. We also provided a tight-binding calculation for \( \omega < \omega_p \) when \( \ell = 1 \).

In contrast to \( \omega > \omega_p \), we found it very difficult to obtain convergence of the photonic band structures below \( \omega_p \) even using a large number of plane waves. This kind of problem was already encountered and discussed in Ref. [105], for a different photonic crystal structure. In agreement with their comments, we can conclude, on the basis of our calculations for \( \omega < \omega_p \), only that there are indications of flat photonic bands, but we have not obtained convergence in our attempts to calculate these using
plane wave expansions. A further study of this frequency region would certainly be of interest.

If we include $N$ plane waves in our expansion, we often find that $2N$ eigenvalues are nonzero and $N$ are zero, as in previous work [195]. The $2N$ bands may correspond to the transverse modes while the $N$ zero bands correspond to the longitudinal modes. We find this behavior without problem for $\omega > \omega_p$, but for $\omega < \omega_p$, it is difficult to draw this conclusion unambiguously because many modes occur at very low frequencies, which might correspond to zero modes if the bands were fully converged.

The fact that the eigenvalues for the $\mathbf{B}$ field in the plane wave expansion method when $\omega < \omega_p$ are accumulated near $\omega = 0$ seems to be related to that there are no TE modes for $\omega < \omega_p$ in an isolated spherical cavity as well as when $kR \ll 1$ and $k'R \ll 1$. As we also see that the $\mathbf{B}$ field results are closer to the tight-binding results in the tight-binding eigenfrequency region, we can roughly reach a conclusion that the plane wave expansion with $\mathbf{B}$ field is better for $\omega < \omega_p$ at least within our calculation results even though we still have a convergence problem here.

Although most of our calculations used only a standard plane wave expansion method, combined with Bloch’s theorem, there are many other band structure calculation methods which could be adapted from conventional electronic band theory and might lead to better convergence for the present problem. Some promising possibilities include the Korringa-Kohn-Rostoker (KKR) method [99, 98, 188], the linear muffin tin orbital (LMTO) method [4, 157], and others. Even in the plane wave method, there are some improved versions, such as the augmented plane wave (APW) method [158, 159, 150] and linearized APW method [5, 97]. It would of interest to compare our results with those obtained by some of these methods in the future.
These methods might solve the convergence difficulties encountered in our results when \( \omega < \omega_p \).

In summary, the convergence problem we encountered for \( \omega < \omega_p \) is similar to that in Ref. [105] for different systems, but the plane wave expansion for 3D metal inverse opals, the single-cavity approach, and the tight-binding method for modes \( \omega < \omega_p \) are all new.
CHAPTER 7

CONCLUSION AND DISCUSSION

Throughout this work, we’ve studied three topics for composite superconductors and two topics for composite dielectrics. Since the intermodulation used an analogy to composite dielectrics to represent the inhomogeneity of superconductors, it stands midway between composite superconductors and composite dielectrics.

In Chapter 2, we’ve seen several evidences for the continuous phase transition in a fully frustrated 3D XY model and found the critical temperature to be $0.681J/k_B$. We also calculated some critical exponents, $\alpha$ of the specific heat, $\nu$ of the helicity modulus, and $\nu$ of the correlation length with various methods. These results can be probed experimentally in a suitable 3D lattice of coupled superconducting grains. Our results give us a single phase transition temperature, but we cannot definitively rule out the possibility of two separate phase transitions. To answer this question we need more numerical studies, especially of the discrete order parameter.

In Chapter 3, we’ve observed a transition from a Mott insulator to a superconductor with increasing $K$ for $\Delta A_{ij} \leq 0.854$ in a disordered 2D superconducting film. For $\Delta A_{ij} > 0.854$, the transition is from a Mott insulator to a Bose glass as $K$ increases. For the superconductor to insulator transition, $K_c$ and $\sigma^*$ increase monotonically with increasing $\Delta A_{ij}$. For the Bose glass to insulator transition, $K_c$ increases, but
\( \sigma^* \) decreases with increasing \( \Delta A_{ij} \). We also calculated the critical values of coupling constant \( K \) and the universal conductivity \( \sigma^* \) at \( K_c \) for several values of \( \Delta A_{ij} \). For disordered films, our model gives a superconductor–insulator transition or a superconductor–Bose-glass transition with increasing \( \Delta A_{ij} \), depending on \( K \).

In Chapter 4, we have presented a general formalism for calculating the intermodulation coefficient and the corresponding intermodulation supercurrent density \( J_{\text{IMD}} \) of an inhomogeneous superconductor. We have also given a simple way to calculate \( J_{\text{IMD}} \) approximately in several geometries. Since such inhomogeneities are known to exist in many of the high-\( T_c \) cuprate superconductors, this formalism is directly relevant for treating an important property of these materials. We find that the resulting \( J_{\text{IMD}} \) is very sensitive to the exact spatial distribution of gaps within the inhomogeneous layer and thus may increase or decrease, depending on the topology. Our calculations show that one way to achieve a large \( J_{\text{IMD}} \) is to have the component with the smaller gap and larger superfluid density embedded in the component with the opposite properties. This appears to be the topology seen in the underdoped BSCCO-2212, which thus may be well suited for a material with a large \( J_{\text{IMD}} \).

In Chapter 5, we could express the electrostatic force between two spheres in simple closed analytic form instead of numerical derivatives. We saw the magnitude of force in the parallel configuration is much greater than that in the perpendicular case at very small separation, but it approaches the dipole-dipole limit at large separation. In addition, the magnitude of force decreases with the increase of frequency and we could see the sign change of force in a slightly conducting host at very small separation. We also checked the validity of \( \ell_{\text{max}} \) included in the matrices for the parallel and perpendicular configurations with the plots of \( F_{\text{av}} \) as a function of \( \omega/\omega_p \).
Our method is applicable to molecular dynamics and magnetorheological fluids just replacing permittivity with permeability. In this work we confined our calculation to only two spheres, but it can be easily extended to three-body or multibody forces.

In Chapter 6, we have used a plane wave expansion for either $E$ or $B$ to calculate the photonic band structures of metallic inverse opals for $\omega > \omega_p$ as well as for $\omega < \omega_p$. We assumed a Drude dielectric function for the metallic component. For $\omega > \omega_p$, we obtained the same results using either field. However, the results for $\omega < \omega_p$ depended both on the number of plane waves used and on whether the expansion was carried out for the $E$ field or $B$ field. We also used a single-cavity approach and a simple tight-binding approach for frequencies $\omega < \omega_p$. Only a few of the plane wave band minima could be understood using this single-cavity approach. We concluded that the plane wave method converged well for $\omega > \omega_p$, but not for $\omega < \omega_p$. We also concluded that, for the cavity sizes studied experimentally, there should be a large number of narrow photonic bands in the range $\omega < \omega_p$. 


[82] J. D. Jackson. *Classical Electrodynamics*, pages 167–169. Wiley, New York, Third edition, 1999. Specifically, when the spheres are moved at fixed potential on the boundaries, the energy $W$ of the system changes. However, the batteries which hold the potential fixed supply twice as much energy to the system as the change in $W$. A discussion of differences between force calculations at fixed potential and fixed charge is in this Ref.


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