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OPTICAL SPECTROSCOPY STUDIES OF BODY CENTERED CUBIC COBALT

AND

GALLIUM ARSENIDE QUANTUM WELL HETEROSTRUCTURES

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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* * * * *

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To my family
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A.1 Top view of (110) plane bcc Co film on a GaAs substrate. The primed axes are obtained by a 45° counter clockwise rotation about z. In this frame, the principal crystal directions [ T0] and [001] lie along the y' and z' respectively. The free surface of the film is at x' = -h and the substrate grows in the direction x' → ∞
The phenomenon of Brillouin light scattering (BLS) was independently and nearly simultaneously discovered by Brillouin and Mandelshtam [1,2]. Material properties are studied by analyzing inelastically scattered light from thermal excitations. Phonons, magnons, plasmons etc. are examples of excitations that scatter light and they provide information on the mechanical, magnetic and electronic properties respectively. BLS can be used to study a diverse range of systems such as liquids, bulk solids and thin film heterostructures. In order to better understand the nature of these excitations, external perturbations like hydrostatic pressure, uniaxial strain, electric and magnetic fields, temperature etc. may be applied to the system under consideration. Some advantages inherent to BLS are its non-destructive nature and the adequacy of small specimen sizes for analysis. Sample dimensions in Brillouin spectroscopy experiments are limited only by the diameter of the focused laser spot (~50 μm), but handling convenience dictates that sample sizes of at least 1mm² are preferred. It is worth noting here that a high sample surface quality is essential for Brillouin spectroscopy because specular reflections must be minimum in order to resolve weak Brillouin signals. Hence, the development of advanced
materials processing techniques like Molecular Beam Epitaxy (MBE) deserve credit for many applications of Brillouin spectroscopy in probing excitations from novel low dimensional heterostructures.

The first project in this thesis involves a measurement of the elastic constants of body centered cubic (bcc) cobalt (Co) from the surface Rayleigh phonon dispersion using Brillouin light scattering. Cobalt, which is a ferromagnetic 3-d transition metal occurs naturally at room temperature as a hexagonal close pack (hcp) structure and transforms into a face centered cubic phase (fcc) at temperatures over 700K. BCC Cobalt is one of the first artificial crystal structures that was stabilized by growing on a lattice matched substrate (GaAs) [3]. It is therefore interesting to study the material properties of this novel synthetic structure. Further, it is known that magnetism plays an important role in influencing elastic properties such as the bulk modulus in 3-d metals [4,5]. Hence there is much theoretical interest in performing first principle calculations to extract the elastic and magnetic properties of 3-d metals [6,7]. The elastic properties of Co, that occurs in various crystal phases therefore provides a natural test-bed to verify the accuracy and validity of various theoretical models. The elastic constants of bcc Co were therefore measured to provide an experimental result to verify the reliability of total energy calculations.

The spin waves in bcc Co were also studied using BLS in order to determine various magnetic parameters such as the exchange, magneto-crystalline anisotropies etc.
of bcc Co [8-11]. The spin wave dispersions reveal that some samples possess no magnetocrystalline anisotropies and instead show large uniaxial anisotropies. A simple model based on magnetoelastic effects that arise from a small film-substrate lattice mismatch, is used to explain the origin of the uniaxial anisotropies [12]. Good agreement is obtained between the anisotropy values measured using Brillouin light scattering and the results obtained from magnetoelasticity calculations. Brillouin scattering and d.c. SQUID measurements also show that the net magnetization of 200Å thick samples are reduced by about 15% from the values measured in thicker samples (~350Å) which are in turn about 10% lower than the theoretical predictions for bulk bcc Co [13]. It has been proposed that arsenic diffusion from the GaAs substrate is responsible for this observed reduction in the magnetization that is more dramatically manifest in thinner films [14].

The second part of this thesis deals with the study of excitons in ultra-thin (15Å - 30Å wide) GaAs/AlGaAs quantum wells using continuous wave (cw) and time resolved photoluminescence (PL) techniques. GaAs quantum wells find important engineering applications in high speed opto-electronic devices. Excitons strongly influence the optical and electronic properties of quantum wells due to an increase in their binding energies under 2-D confinement. In very thin quantum wells, rough hetero-interfaces scatter electrons and trap excitons. Hence the quality of the interface is also an important factor in determining the performance characteristics of these devices. A good understanding of the role of hetero-interface ordering on the lifetimes and linewidths of excitonic PL from
quantum wells is therefore highly desirable [15]. Growth interruption is a standard technique employed during MBE to minimize the effects of interface roughness and produce smooth interfaces [16,17]. However, this procedure creates large island-like regions in the wells that vary in width by about one atomic monolayer. Excitons in growth interrupted quantum wells are localized at islands and this effect is manifest as a fine structure in the excitonic PL [16-18].

Past research on excitonic PL from growth interrupted quantum wells involved a continuous wave study and some limited time resolved studies on exciton lifetimes [18-20]. This work involves a detailed temperature dependent cw and time resolved study of excitons, localized at islands of different well width in growth interrupted single GaAs/AlGaAs quantum wells. The cw results show doublets in the exciton PL that corresponds to excitons localized in regions of different well width. The doublet oscillator strength switches from the low energy to the high energy side with increasing temperature. The time resolved data reveal that the excitons from the doublet have nearly the same lifetimes that scale linearly with temperatures under low power excitation ($n \sim 10^{10}$ cm$^{-2}$). These results are interpreted in terms of the dimensions of the island diameter ($d$) as compared to the exciton diffusion length ($L_D$). In the regime $d \gg L_D$, excitons from different island regions that vary in width, do not couple through diffusion. Hence exciton decays have nearly identical lifetimes. When $d \sim L_D$, the two decay channels are strongly coupled through diffusion and the higher energy PL has a shorter lifetime than the lower
energy one [18]. Exciton lifetimes obtained from time resolved PL measurements enables us to place limits on the island sizes. PL spectroscopy therefore provides a non-destructive method to probe hetero-interface ordering in quantum wells.

The dynamics of exciton lifetimes and decay lineshapes are examined under high carrier densities \( (n \text{ upto } 10^{12} \text{ cm}^{-2}) \) in ultra-thin GaAs/AlGaAs single quantum wells. At low carrier excitation densities \( (n \sim 10^9-10^{10} \text{ cm}^{-2}) \), the excitonic PL exhibits a mono-exponential decay profile and the predicted linear dependence of lifetimes on temperatures is observed up to 100K. At intermediate carrier concentrations \( (n \sim 10^{11} \text{ cm}^{-2}) \), we observed strong deviations from the mono-exponential decay that suggest a cooling behavior. At peak carrier densities achieved in the experiments \( (n \sim 10^{12} \text{ cm}^{-2}) \), which exceeds the critical Mott density for the existence of exciton bound states, the PL decay occurs in 3 stages. A new mono-exponential profile appears at the onset of the PL decay followed by the cooling profile observed previously. The last segment of the decay again appears as a mono-exponential similar to the low excitation density case. The lifetime of the initial mono-exponential is independent of temperature while the cooling segment relaxes to a mono-exponential as the sample temperature increases. These complex decay patterns observed at high carrier densities possibly require many body effects to completely understand the data. Since very little theory is available on this subject, we speculate on the mechanisms underlying the decay patterns based on their response to temperature.
References


CHAPTER II

Fabry-Perot Instrumentation

This chapter discusses details of Brillouin spectroscopy instrumentation used in the elasticity and magnetism studies on body centered cubic cobalt thin films. The Brillouin spectrometer is based on a Sandercock type tandem Fabry-Perot Interferometer (FPI) and was set-up in this laboratory by Dr. J.M. Karanikas [1]. Details of it’s construction are provided in reference [1]. The discussion below is limited to the following topics: theory of operation, factors influencing performance and a description of the experimental setup.

1 Introduction

In order to appreciate the difficulties encountered in Brillouin spectroscopy instrumentation, it is instructive to compare it on the energy scale with Raman spectroscopy. While grating spectrometers are adequate for Raman research, the requirements of Brillouin spectroscopy are more demanding.

Brillouin light scattering (BLS) is an inelastic light scattering process that proceeds via mechanisms similar to Raman scattering [2]. However, while Raman scattering examines excitations with energies typically > 20 cm$^{-1}$, BLS probes extremely low energy excitations in the range of 0.1 - 1 cm$^{-1}$. Hence, while instrument resolutions of the order
of 1 cm\(^{-1}\) are adequate for Raman spectroscopy, BLS require resolutions better than 0.005 cm\(^{-1}\). Raman spectrometers have an instrument linewidth of \(\sim 0.2\) cm\(^{-1}\) while for BLS we require linewidths < 0.2 GHz (1 cm\(^{-1}\) = 30 GHz). The proximity of the weak Brillouin signal to the strong elastic laser line (high contrast needed, \(10^6\) to \(10^{10}\)) and the need for high resolution imposes severe demands on the BLS instrumentation. Optical heterodyne, grating and Fourier transform spectrometers all fail to satisfy either the resolution or the contrast requirements of Brillouin spectroscopy [3-5]. The Fabry-Perot interferometer (FPI) was shown to be the only instrument possessing the potential for Brillouin spectroscopy applications. Even so, the first single pass Fabry-Perot spectrometer did not possess the contrast required for Brillouin spectroscopy [6,7]. The tandem multi-pass FPI designed by Sandercock circumvented previous design problems [8-10]. This development lead to an explosive growth in the field of Brillouin spectroscopy which was otherwise languishing from lack of suitable instrumentation.

2 Fabry-Perot Interferometry

A Fabry-Perot Interferometer (FPI) consists of two optically flat surfaces with high reflectivity dielectric coatings that form a resonant optical cavity. Figure 2.1 shows a FPI in a single pass configuration. A detailed analysis of this device is available in books and review articles [5,11,12]. Transmission maxima occur when the round trip resonance condition given by \(n\lambda = 2L\) is satisfied, where \(L\) is the optical length of the cavity, \(\lambda\) is the
wavelength of light and \( n \) is an integer. The ratio of the transmitted to incident beam intensity depends upon the optical cavity length (plate spacing) \( L \) and its reflectivity \( R \).

\[
\frac{I_t}{I_0} = \left( \frac{T}{1-R} \right)^2 \frac{1}{1+\left(\frac{2F}{\pi}\right)^2 \sin^2 \left(\frac{2\pi L}{\lambda}\right)}
\]  

(2.1)

\( T \) is the transmission coefficient and \( R+T+A = 1 \) where \( A \) is the absorption coefficient. \( F \) is the overall finesse of the spectrometer defined by

\[
F = \frac{FSR}{FWHM}
\]

(2.2)

where FSR (free spectral range) is the spacing between adjacent resonances and FWHM is the full width at half maximum or the linewidth of the transmitted light. The FSR is equal to \( c/2L \) where \( c \) is the velocity of light. Hence the bandwidth of the interferometer can be varied through \( L \). From equation (2.1), the positions of the transmission resonances can also be controlled by through \( L \). Hence, the FPI can be used as a high resolution tunable filter where only a particular spectral component is transmitted at a given plate spacing.

Figure 2.2 shows how finesse affects the lineshape of the transmission function \( I_t \). Transmission peaks appear like delta functions for large \( F \) and are broad when \( F \) is small. The main factors contributing to the instrumental finesse are the plate reflectivity finesse, \((F_R)\) and the surface flatness finesse \((F_S)\). Secondary factors degrading \( F \) are diffraction losses and output pinhole losses. The reflectivity finesse is given by

\[
F_R = \frac{\pi \sqrt{R}}{(1-R)}
\]  

(2.3)
The FPI plate reflectivity in our system is \( R = 0.94 \) at 5145 Å which gives \( F_R = 50 \). Roughness of the optical flats produces specular reflections that broaden the transmission function and thus reduce \( F \). The surface flatness finesse is given by the expression

\[
F_S = \frac{m}{2}
\]

where \( m \) is the flatness of the optical plates expressed in units of \( \lambda \). The FPI plates used in our setup were flat within \( \lambda/150 \) which gives \( F_S = 75 \). The overall instrument finesse expressed in terms of the above quantities is [12,13]

\[
\frac{1}{F^2} = \sum_{i=1}^{n} \frac{1}{F_i^2}
\]

where the index \( i \) refers to the different contributions to finesse. Taking \( F_R \) and \( F_S \) into account we get \( F = 40 \). The overall finesse will reduce to 30 or less if diffraction and pinhole contributions are included. In order to obtain high resolution, (i.e. small FWHM) we require a large instrument finesse. For example, a plate spacing \( L = 6 \) mm corresponds to a FSR of 25 GHz. Thus in order to obtain a FWHM of 0.2 GHz, we require an instrument finesse of 125 whereas a single pass configuration yields a finesse of only 30.

The instrument contrast is another figure of merit that is critical for Brillouin spectroscopy. This is the ratio of the transmission intensity maximum to the intensity minimum and it depends only on \( F \).

\[
C = 1 + \frac{4}{\pi^2} F^2
\]
When $R = 0.94$, we obtain $C = 350$. The above contrast is clearly inadequate to extract Brillouin signals that are $10^6 - 10^{10}$ times weaker than the elastic laser line. Hence, neither finesse nor contrast in a single pass FPI configuration meet the minimum requirements for Brillouin spectroscopy.

The contrast and finesse of a FPI improve dramatically on multi-passing. Corner cubes serve as retro-reflectors to multi-pass the input beam (figure 2.3) and the resulting expressions for finesse and contrast are

$$ F_n = \frac{F}{\sqrt{2^n - 1}} \quad \text{and} \quad C_n = C^n \quad (2.7) $$

where $n$ is the number of passes used and $F$, $C$ are the single pass finesse and contrast defined previously. A typical multi-pass configuration with $n = 6$ yields acceptable values of finesse ($F_n = 250$) and contrast ($C_n = 2 \times 10^{15}$).

Light scattered from a sample is spectrally analyzed by varying the FPI plate spacing continuously over a distance of a few microns. The spectral components consisting of elastic and inelastic signals are resolved because, for a given plate spacing $d_0$ only a particular wavelength component $\lambda_0$ is transmitted (eqn. 2.1). When several successive elastic orders are transmitted during a scan, it is difficult to determine which elastic order a particular Brillouin peak belongs to. The elastic order $n_0$ associated with a plate spacing $L_0$ and wavelength $\lambda_0$ is known because of the relation $2L_0 = n_0 \lambda_0$. An
inelastic peak with wavelength \( \lambda_i = \lambda_0 + \delta \lambda \) is transmitted at plate spacing \( L_i \) only if transmission resonance condition

\[
2L_i = (n_0 + n_i)(\lambda_0 + \delta \lambda) \quad \text{or} \quad \lambda = \frac{L_1 - L_0 - \left( \frac{n_1 \lambda_0}{2} \right)}{n_0 + n_1} \tag{2.8}
\]

is satisfied. Thus, the unknown transmission order \( n_i \) is obtained by repeating the experiment at two different plate spacings. Assigning an inelastic peak to a particular elastic order is not difficult if only one or two non-overlapping inelastic peaks are present. However, when interpreting complex spectra involving multiple peaks over a wide range of energies, inelastic peaks from different orders may overlap. This would make it difficult to unambiguously assign an inelastic signal to it’s elastic order.

The above problem is avoided by using a tandem FPI where two optical cavities oscillate in tandem [8-10]. The plate spacings of the two cavities are selected so to filter out all but one elastic order. Hence Brillouin signals passing through both cavities must belong to that transmitted order. The resonance conditions for plate spacings \( L_1 \) and \( L_2 \) is

\[
2L_1 = n_1 \lambda \quad \text{and} \quad 2L_2 = n_2 \lambda. \tag{2.9}
\]

The response function of a tandem interferometer is a convolution of it’s individual interferometer response functions (figure 2.4). An inelastic signal with wavelength shift \( \delta \lambda \) from the elastic line of wavelength \( \lambda \), must simultaneously satisfy the relations

\[
\frac{\Delta L_1}{L_1} = \frac{\delta \lambda}{\lambda} \quad \text{and} \quad \frac{\Delta L_2}{L_2} = \frac{\delta \lambda}{\lambda} \tag{2.10}
\]
in order to pass through both interferometers. Here \( \Delta L_1 \) and \( \Delta L_2 \) are the scan amplitudes of the plates with spacings \( L_1 \) and \( L_2 \) respectively. From equations 2.9 and 2.10, the relative late spacings must also satisfy the equation

\[
\frac{L_1}{L_2} = \frac{\Delta L_1}{\Delta L_2} = \frac{n_1}{n_2}
\] (2.11)

The tandem FPI used in a multi-pass configuration therefore satisfies all the conditions required for Brillouin spectroscopy, i.e. high resolution, high contrast and ease of spectral identification. However, alignment of the two interferometers is now a non-trivial task. The two sets of interferometer plates must not only must be maintained parallel during the scan but their scan must be dynamically synchronized so that only one elastic order passes through. Further, parallelness of both sets of interferometer plates must be maintained over long periods of time (2-3 hours). Initial attempts at pressure scanning a tandem FPI unit failed due to lack of long term stability [16,17]. The Sandercock model overcomes the problems of pressure scanning by using a deformable parallelogram.

Figure 2.5 shows the top view of the Sandercock tandem FPI. A translational stage holds two plates, one for each cavity, with the normal to the cavity faces intersecting at an angle \( \theta = 20.4^\circ \). Two more plates held by rigid mounts complete the other halves of the optical cavities. The plate spacing is set to \( L \) for one interferometer and \( L\cos\theta \) for the other. Small oscillations of the two cavities about their set spacings automatically satisfies equation 2.11. Rigid, tilt free translation during a scan is attained using a deformable
parallelogram that rides atop a roller translational stage (figure 2.6). This stage uses precision stiffened steel flats as guides that achieve near collinear displacements over distances of the order of 1 cm. Scans are actuated by a piezo-electric transducer mounted on the translational stages and are driven by a D/A converter on the interferometer controller. The D/A converter generates a sawtooth wave pattern that expands the cavity and data are collected at this time. A single scan takes about 1 second and several thousand scans are required for a high quality spectrum. The scan displacement is measured using a small air gap capacitor. During the scans, a feedback control loop provides dynamic stabilization of the interferometer and maintains plate parallelness. This is done by applying corrective voltages to three sets of piezo-electric stacks which define a plane, mounted on the FPI plates of the scanning stage. The above arrangement provides a synchronized, linear, tilt free scan with <5Å lateral drift [18].

3 Experimental Setup

Figure 2.7 shows a schematic layout of the experimental setup. An Ar ion laser is used as an excitation source. The intrinsic laser linewidth is too wide for BLS experiments and is therefore narrowed down to 100 MHz using a temperature stabilized intra-cavity etalon. An acousto-optic modulator is inserted in the optical path of the laser to attenuate the laser beam and protect the sensitive GaAs photomultiplier detector. Experiments are performed in back-scattering where collimating optics focuses laser light on the sample
and collects the scattered radiation for analysis. The scattered light, consisting of elastic and inelastic components is collimated, filtered through a pinhole and then directed normally on the first FPI. A multi-pass (6 passes) arrangement through the tandem is implemented using a large rear mirror, a corner cube and a cat-eye. The output beam is directed onto a 3 nm interference filter (centered at 5145Å) that suppresses laser plasma lines and stray specularly scattered radiation. The final beam is again filtered before detection. A thermo-electrically cooled GaAs photomultiplier is used to detect signals. A multi-channel scalar card along with data acquisition software collects data during scans and stores them in a computer. The FPI and GaAs detector are enclosed within a light tight box and all lights are turned off during data acquisition. The typical integration time to acquire a Brillouin spectrum is 2-3 hours. Due to the delicate nature of the apparatus, the entire system is placed on a floating air table in order to isolate vibrations. The interferometer stability is sensitive to temperature fluctuations. Hence, care is taken to maintain thermal stability (< ± 0.5° C) within the FPI enclosure.
Figure 2.1: Basic FPI arrangement consists of two plane parallel optical surfaces $S_1$ and $S_2$. The surfaces are flat within $\lambda/150$ and are coated with a high reflectivity dielectric coating. A collimating lens ($L_1$) collects light from the sample and a second lens $L_2$ focuses the interferometer output into a pinhole. The diameter of the pinhole is a carefully balanced choice between resolution and light collection light efficiency (reference [12], pg. 98).

Figure 2.2: Interferometer transmission function (equation 2.1) for a different values of finesse, $F$. Top graph is plotted on a log scale while bottom one is linear (reference [12], pg. 100).
Figure 2.3: 1. FPI in a multi-pass configuration using a corner cube, in order to enhance contrast and finesse (reference [16], pg. 374). 2. Fabry-Perot used as a tunable filter. As plate spacing is varied only a particular spectral component is transmitted for a given plate spacing. Top portion of figure shows FPI being scanned and separated spectral components are shown below. The strong peaks are the elastic orders while the weak peaks are the inelastic Brillouin signals (reference [14], pg. 372).
Figure 2.4: Elimination of multiple elastic orders by using two FPI's in tandem with unequal plate spacings. Transmission function is limited to a single elastic orders with weak ghosts arising from a weak overlap of the adjacent orders (reference [15], pg. 184).

Figure 2.5: A translational stage of a tandem FPI designed to synchronize both stages during a scan. When plate spacing for FPI₁ is \( L \), the corresponding spacing for FPI₂ is set to \( L \cos \theta \) (reference [15], pg. 186).
Figure 2.6: Scanning stage of FPI built using a deformable parallelogram rides atop a roller translational stage. The capacitance measurement circuit and D/A converter that drives the scanning transducer are also shown (reference [15], pg. 185).
Figure 2.7: Schematic diagram of Brillouin experimental setup (reference [1]).
References


CHAPTER III

Acoustic Waves in Solids

The first part of this chapter presents the theory of acoustic waves in solids based on solving the classical wave with appropriate boundary conditions. The solutions give rise to bulk and surface-like excitations. In supported media, a number of guided waves may exist depending upon the elastic properties of the film and substrate. The properties of various elastic waves are correlated to the elastic properties of the media. The second part of the chapter discusses light scattering mechanisms for acoustic waves in solids.

1 Acoustic Waves In Thick Films

Acoustic waves constitute any time varying mechanical disturbance in a medium. Lord Rayleigh first demonstrated that waves exist at surfaces of all solids [1]. This result had maximum relevance to geology and much of the seminal work on acoustic waves in solids was therefore done by geologists [2-5]. A classical macroscopic description of wave propagation in solids is valid because the wavelength of excitations are many orders of magnitude greater than inter-atomic spacings. The macroscopic factors that govern the properties of elastic waves in a medium are the elastic ‘spring’ constants and the density
of the medium. The dispersion properties of various acoustic waves are sensitive to different elastic constants. Hence studies of acoustic wave dispersion provide a direct measure of the elastic constants they are sensitive to. The equation of motion in the absence of piezo-electric effects and external forces is given by

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \frac{\partial T_{iy}}{\partial x_i} \quad (i, j = 1, 2, 3)$$

(3.1)

in which $\rho$ is the density of the medium, $T_{ij}$'s are elements of the stress tensor, $u_i$'s are the displacement fields. In the linear regime, the strain field $S$ produced by these displacements is given by

$$S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$

(3.2)

Non-linear terms in strain can be neglected for strains $< 10^{-4}$ [6]. Hooke's law provides a relation between stress $T$ and the strain $S$

$$T_{ij} = C_{ijkl} S_{kl}$$

(3.3)

where $C_{ijkl}$'s are the elastic constants. The equation of motion can be rewritten using the above relations as

$$\rho \frac{\partial^2 u_j}{\partial t^2} = C_{ijkl} \frac{\partial^2 u_k}{\partial x_i \partial x_l} \quad (i, j, k, l = 1, 2, 3)$$

(3.4)

In an unbounded semi-infinite medium plane wave solutions satisfy the wave equation

$$u_i = u_{i0} \exp[i(k \cdot r - wt)] \quad (i = 1, 2, 3)$$

(3.5)
where $k$ is the wave-vector and $\omega$ is the frequency of the plane wave. The wave-vector $k = (k_1 x + k_2 y + k_3 z)$ where $k_1$'s are components of the wave-vector and $\lambda$'s are eigenvalues. When (3.5) is substituted into (3.4), we obtain an eigenvalue equation that has solutions only when the determinant given by

$$|C_{ijkl} l_j l_k - \rho \nu^2| = 0$$

(3.6)

This determinant produces a secular equation cubic in the eigenvalue $\nu^2$ for propagation along arbitrary crystal directions. For bulk-like modes all eigenvalues ($\lambda$'s) are real and the waves radiate into the volume of the film. In isotropic solids, waves decouple for different polarizations and familiar solutions for bulk shear and longitudinal modes with velocities $\sqrt{\frac{C_{44}}{\rho}}$ and $\sqrt{\frac{C_{11}}{\rho}}$ respectively are obtained. These expressions become more complex and depends on combinations of various elastic constants when crystals with lower symmetry or arbitrary propagation directions are considered. Mode separation into pure shear and longitudinal waves occurs only along specific crystal directions [6-9].

When the wave-vector component normal to the film plane becomes complex, the amplitude of the wave becomes strongly localized at the surface. This wave, known as a Rayleigh or surface wave, exists on all surfaces and it's amplitude decays exponentially into the bulk. Rayleigh waves in unsupported layers are non-dispersive and their velocities ($v_R$) are always less than the shear velocity of the medium. An approximate expression for $v_R$ in terms of the transverse ($v_T$) and longitudinal ($v_L$) bulk velocities is given by [10]
\[
\frac{v_R}{v_T} = \frac{0.87 + 1.12\sigma}{1 + \sigma} \quad \text{and} \quad \sigma = \frac{1 - 2\left(\frac{v_T}{v_L}\right)^2}{2\left(1 - \left(\frac{v_T}{v_L}\right)^2\right)^2}
\]  
(3.7)

where \(\sigma\) is the Poisson’s ratio (0 < \(\sigma\) < 0.5). Figure 3.1 shows the dependence of the Rayleigh wave velocity on \(v_T\) and \(v_L\) in an isotropic medium. Hence \(v_R\) is sensitive to the elastic constants \(c_{11}\) and \(c_{44}\). In all cases other than isotropic solids the Rayleigh wave velocity can only be calculated numerically. Figure 3.2 shows the Rayleigh wave amplitude in an isotropic unsupported layer. The \(u_1\) and \(u_3\) displacement components lie in the sagittal plane and decay exponentially within a few wavelengths into the films.

2 Waves in Supported Films

2.1 Rayleigh Modes

Supported films permit a rich spectrum of guided modes such as Lamb waves, Love modes, Sezawa modes and in special cases, interface modes also known as Stoneley modes [2-5]. Figure 3.3 shows a schematic diagram of a supported film of thickness \(h\) on a semi-infinite substrate \((x_3 \rightarrow -\infty)\). The film-substrate interface is at \(x_3 = 0\). The sagittal plane is the vertical plane defined by the normal to the film and the propagation direction. The horizontal in-plane lines are perpendicular to the sagittal plane. When a wave propagates along arbitrary directions, the normal and in-plane polarizations are coupled and the resulting wave is known as a generalized Lamb wave. Along directions of high
crystal symmetry, the waves decouple into in-plane and out-of-plane polarizations known as Love modes [2] and Rayleigh modes respectively [1].

In a supported film, the wave equation is solved separately for the film and the substrate (Appendix A) and independent secular equations (3.6) are obtained for the two media. Once the eigenvalues are computed, displacement fields in the film and substrate are constructed as superpositions of plane partial waves using the eigenvalues for that medium. For surface-like modes where the eigenvalues corresponding to $k_i$'s normal to the film are complex, we obtain six partial wave solutions for the film and only three for the substrate. This is because the substrate is an infinite medium and partial waves corresponding to diverging solutions are neglected. Since the sagittal plane and in-plane displacements decouple for propagation along symmetry directions, the corresponding secular equation decouples into fourth order and second order polynomial equations (in the eigenvalue $l_i$) corresponding to the Rayleigh and Love modes respectively.

The partial wave solutions for the films and substrate satisfy the mechanical boundary conditions across the interface and at the free surface of the film. The displacement is continuous across the interface at $x_3 = 0$.

$$u_i^f = u_i^s \quad (i = 1, 2, 3) \quad (3.8(a))$$

The stress fields are continuous across the interface at $x_3 = 0$.

$$T^f \cdot n = T^s \cdot n \quad (3.8(b))$$
Lastly, the free surface of the film is exposed to air or vacuum. Since the gaseous medium above cannot a tangential stress, the upper surface of the film must be traction free. Thus

\[ T_f \cdot \hat{n} = 0 \text{ at } x_3 = h \]  \hspace{1cm} (3.8(c))

The superscripts \( f \) and \( s \) refer to the film and substrate respectively and \( \hat{n} \) is a unit normal to the interface or film surface. When examining Rayleigh waves in supported films we obtain four partial wave solutions in the film and two in the substrate. Hence the number of useful boundary conditions from equation (3.8) reduce to six. The net displacements in the film and substrate are superpositions of the partial waves using a (yet unknown) set of amplitudes \( f_i \)'s.

These six boundary conditions expressed in the form of a homogenous set of equations possess a solution only if their corresponding determinant \( |M_{ij}| = 0 \).

\[ \sum_{j=1}^{6} M_{ij} f_j = 0 \hspace{1cm} (i, j = 1..6) \]  \hspace{1cm} (3.9)

\[ |M_{ij}| = 0 \]  \hspace{1cm} (3.10)

The lowest value of the phase velocity that satisfies this condition is the Rayleigh wave solution. This wave exists in all supported films regardless of any material parameters. A detailed procedure for calculating the Rayleigh dispersion in anisotropic supported films is given in appendix A [1,11].

The main difference between Rayleigh waves in bulk media and those in supported films is that the Rayleigh wave is dispersive in the latter. The details of this dispersion
behavior are intimately connected with the transverse bulk velocities of the film and substrate.

1. Film transverse velocity greater than substrate transverse velocity ($v_{t}^f > v_{t}^s$):

When $v_{t}^f > v_{t}^s$ the film is said to stiffen the substrate. This happens when the slope of the plot $\frac{c_{44}^f}{c_{44}^s}$ vs $\frac{\rho_s^f}{\rho_s^s}$ exceeds unity. For material combinations of this type, the dispersion curve starts at the substrate Rayleigh velocity for $kh = 0$ and increases to the substrate shear velocity $v_{T}^f$ at some finite $kh$ (figure 3.4.1). Only one surface mode is possible here and it gets delocalized once the substrate shear velocity is reached. There is no simple analytical expression to evaluate the cut-off value of $kh$.

2. Film transverse velocity less than substrate transverse velocity ($v_{t}^f < v_{t}^s$):

When $v_{t}^f < v_{t}^s$, the film loads the substrate and this condition arises when the slope of $\frac{c_{44}^f}{c_{44}^s}$ vs $\frac{\rho_s^f}{\rho_s^s}$ is less than unity. Figure 3.4.2 shows an example of a Rayleigh wave on a film that loads a substrate for isotropic parameters. At $kh = 0$, the Rayleigh wave takes the substrate Rayleigh wave velocity. The mode velocity decreases with increasing $kh$ and when $kh >> 1$, it becomes asymptotic to the Rayleigh wave velocity of the free standing film. An important feature here is the presence of an unlimited number of guided modes. The higher order Rayleigh modes (i.e. phase velocities that yield $|M_i| = 0$, known as Sezawa modes) have low frequency cut-offs when the phase velocity equals the substrate
shear velocity. These modes retain their surface like character until their velocities approach close to the substrate shear velocity, after which they get delocalized and radiate into the bulk. All the higher order Rayleigh modes velocities have high frequency asymptotes equal to the film shear velocity ($v_T^f$).

Figure 3.5 shows the $u_1$ and $u_3$ (sagittal plane) displacement amplitudes for the third Rayleigh mode ($R_3$ of figure 3.4.2). The low frequency cut-off for this mode occurs at $kh = 1.14$ for the choice of parameters used. When $kh$ is slightly greater than cut-off, the normal displacement amplitude $u_3$ penetrates deep into the substrate. The penetration depth of $u_3$ is now large compared to the wavelength or the penetration depth of $u_1$, as illustrated when $kh = 1.2$. This behavior characterizes a transition from a surface-like to a bulk-like mode. At large $kh$, the sinusoidal spatial dependence in the layer is manifest with the two displacement components being in approximate phase quadrature.

2.2 Stoneley Modes

The previous discussion on Rayleigh modes considered cases where the shear velocities of the substrate and film were considerably different. There exists a special sagittal plane excitation (known as the Stoneley mode) localized at the interface of two half-spaces welded together (i.e. when $h \to \infty$ in figure 3.3) over a limited range of the parameters $c_{44}$ and $\rho$ for the film and substrate [13]. The Stoneley mode occurs when $v_T^f = v_T^s$ and is called an interface mode because it’s amplitude decays exponentially into the bulk of both
media [4,13-15]. The velocity and displacements of the Stoneley wave can be determined by eliminating the growing eigenvalues solutions for the film and the boundary conditions corresponding to a stress free top surface, from the Rayleigh mode analysis. This mode is dispersionless because both media are infinitely thick and hence there is no characteristic length scale in the medium. The Stoneley wave velocity is lower than the shear velocities of both constituent media. It exists at all solid-liquid interfaces but only in very special cases for solid-solid interfaces (ex. Al on W). The relation between a Stoneley mode and a Rayleigh wave can be seen by considering the behavior of the Rayleigh wave as a function of film thickness in a pair of materials capable of supporting a Stoneley mode. Figure 3.6 shows the case of W on Al where the lowest order Rayleigh mode asymptotically becomes the Stoneley mode and for Al on W where the second order Rayleigh mode is asymptotic to the Stoneley mode. Figure 3.7 shows the displacement components of the Stoneley wave clearly indicating it’s interface character.

2.3 Love Modes

Love modes involve displacements perpendicular to the sagittal plane [2,9]. In an isotropic supported film or for special propagation directions in an anisotropic medium, the in-plane and sagittal plane dispersions decouple. The secular determinant correspondingly factors into fourth and second order equations, the latter of which gives the Love mode solutions to the wave equation. Hence there are two partial wave comprising the displacement field
in the film and only one in the substrate. Therefore there are only three boundary
conditions applicable for calculating the Love mode dispersion. The Love mode dispersion
in an isotropic supported film is given by [16]

$$\tan(kh)[(\frac{v}{v_T^f})^2 - 1]^{1/2} = r^{-1} \frac{[1 - (\frac{v}{v_T^f})^2]^{1/2}}{[(\frac{v}{v_T^s})^2 - 1]^{1/2}}$$

(3.11)

where \( r = c^f_{44} / c^s_{44} \). Real solutions for the Love mode exist only if \( v_T^f < v_T^s \). Due to the
periodic nature of the tangent function in the dispersion, multiple solutions exist for a
given set of material parameters provided \( v_T^f < v_T^s \) is satisfied. The velocity of the first of
these modes becomes the substrate shear velocity when \( kh \to 0 \). When the film loads the
substrate, higher order Love modes all have a low frequency cut-off when the phase
velocity of the wave equals the substrate shear velocity. As \( kh \to \infty \), the Love mode
velocities become non-dispersive and asymptotic to the film shear velocity. The particle
displacements decay exponentially into the substrate and oscillate in the film.

3 Light Scattering by Acoustic Waves

Brillouin light scattering by acoustic phonons proceeds via two distinct mechanisms. The
first is the elasto-optic (EO) process where thermal excitations in a medium produce
fluctuations in the dielectric susceptibility that scatter light [17]. This process is similar to
Raman scattering from optical phonons where the susceptibility tensor is expanded in
terms of the normal modes of the lattice vibrations. The EO mechanism is important in transparent solids and is largely responsible for the scattering cross sections of bulk phonons. In the case of surface phonons where the phonon amplitude is localized near the surface, the EO contribution to the total scattering contribution is usually small.

In opaque materials where the penetration depth of light is limited (few 100Å’s), scattering occurs primarily off surface phonons. A phonon traveling on the surface of the solid acts like a dynamical grating that ripples the surface. These surface ripples produce large variations in the susceptibility tensor and scatter light. This mechanism is known as ripple scattering and is the dominant process for scattering from surface waves in metals and other opaque media [18].

There are two general macroscopic approaches to calculating the phonon scattering cross sections due to ripple and EO contributions. The first approach uses a Green’s function formalism and was first introduced by Maraduddin, Loudon and others [19-22]. The second method involves solving Maxwell’s equations in a perturbative scheme, expanding fields to first order in fluctuations in the dielectric tensor, and applying suitable boundary conditions [23-28]. The elasto-optic contribution in bulk modes can be used to determine the elasto-optic constants responsible for scattering where the latter are treated as fitting parameters [27,28]. In transparent supported films strong interference effects have been observed between light scattering from the free surface and the interface [29,30], which provides a rigorous test of the theoretical calculations of scattering
intensities. The theory has recently been extended to study guided waves in tri-layered systems comprising of SiO$_2$ embedded in Si [31,32].

### 3.1 Scattering Cross Section Calculation

The work of Marvin et al is described below in order to review Brillouin light scattering from phonon in unsupported films [26-28]. The medium lies in the $xy$ plane for $z < 0$. Phonon induced polarization is expressed in terms of fluctuations in the dielectric tensor

$$\varepsilon_{ij}^m(r,t) = \varepsilon_{ij}^0 + \delta\varepsilon_{ij}(r,t)$$  \hspace{1cm} (3.12)

where $\varepsilon^0$ is the dielectric tensor in the absence of phonons and $\delta\varepsilon$ represents fluctuations due to phonons. When $\delta\varepsilon$ is expanded in terms of the phonon normal co-ordinates we get

$$\delta\varepsilon_{ij}(r,t) = \sum_{q,n} \delta\varepsilon_{ij}^n(z,q) \exp[i(q.r - \Omega_n t)]$$  \hspace{1cm} (3.13)

where $q$ is the phonon momentum with frequency $\Omega_n$ and $n$ is its degeneracy. The phonon wave-vector $q$ can correspond to either surface or bulk modes. For surface modes there is only an in-plane momentum involved, $q_\parallel$. Equation 3.12 describes the EO contribution to the scattered light arising from strain induced in the medium by vibrational modes. The relation between strain and modulation in the dielectric tensor is

$$\delta\varepsilon_{ij}(r,t) = k_{ijkl}S_{kl}(r,t)$$  \hspace{1cm} (3.14)

$$S_{ij}(r,t) = \frac{1}{2}(\frac{\partial u_i(r,t)}{\partial x_j} + \frac{\partial u_j(r,t)}{\partial x_i})$$
where $k_{ijl}$'s ($S_{ij}$'s) are elements of the elasto-optic (strain) tensor and $u(r,t)$'s are the phonon displacement fields. In order to include ripple effects the medium is defined as the half space $z < u_z(R, z = 0, t)$, where $u_z$ is the normal phonon displacement field that ripples the film surface at $z = 0$. The elasto-optic and ripple effects can simultaneously be included in $\delta e$ as follows:

$$
\varepsilon_y(r, t) = \delta_y \theta [z - u_z(R, 0, t)] + \varepsilon^{in} \theta [u_z(R, 0, t) - z]
$$

$$
\theta (z) = \begin{cases} 
1 & (z > 0) \\
0 & (z < 0)
\end{cases}
$$

Maxwell's equations are solved to first order in $\delta e$ to calculate all the fields in and out of the medium. Since $\delta e$ is expanded in terms of the phonon displacements (equation 3.13, 3.14), the electric and magnetic field amplitudes expanded to first order in $\delta e$ depend on the different phonon displacements in the medium. The phonon displacements can be calculated for different excitations using the procedure outlined in the previous section.

For p-polarized incident light which is the typical scattering geometry for Brillouin experiments, solutions to the magnetic field $B(r,t)$ are obtained using

$$
\nabla \times E + \frac{1}{c} \frac{\partial B}{\partial t} = 0, \quad \nabla \cdot B = 0 \quad and \quad \nabla \times B - \frac{\varepsilon}{c} \frac{\partial E}{\partial t} = 0
$$

which gives

$$
\{ \nabla \times [\varepsilon^{-1} \nabla \times] + \frac{1}{c^2} (\frac{\partial^2}{\partial t^2}) \} B = 0
$$

(3.16)
The relevant boundary conditions of this problem are the continuity of the normal and tangential component of the magnetic field. The Poynting vector is calculated by computing the thermal average of $E \times B$ in order to obtain the scattering cross section

$$S(r, t) = \frac{c}{8\pi} \langle \text{Re}(E(r, t) \times B^* (r, t)) \rangle$$

(3.17)

where $\langle \rangle$ indicates thermal average. The differential scattering cross section is simply a projection of the Poynting vector on the $z$ axis. The expressions for angular dependence of scattering cross sections provide useful insights for Brillouin experiments [27].

$$\left( \frac{d^2 \sigma}{d\Omega_n d\omega} \right)_{s \rightarrow s} \sim \cos^3 \theta_0 \left| 1 + \alpha \sin \theta_0 \right|^2$$

$$\left( \frac{d^2 \sigma}{d\Omega_n d\omega} \right)_{p \rightarrow p} \sim \frac{\cos^3 \theta_0}{(\cos \theta_0 + \varepsilon_0^{-1/2})^4} \left| 1 + \beta \sin \theta_0 + \sin^2 \theta_0 \right|^2$$

(3.18)

$$\left( \frac{d^2 \sigma}{d\Omega_n d\omega} \right)_{p \rightarrow s} \sim \cos \theta_0 |\gamma \sin \theta_0|^2 \quad \alpha, \beta, \gamma \sim \varepsilon_0^{-3/2}$$

The above expressions are valid for back scattering with $\theta_i = -\theta$, which is the most commonly used geometry in Brillouin experiments. At grazing incidence the p-p geometry has the largest cross section. Hence Brillouin experiments in a back-scattering geometry are performed using p-polarized light. Detailed expressions for scattering amplitudes are provided in reference [28]. The essential results of the scattering cross section calculations for the ripple and EO contributions are as follows:
1. Polarized (p-p and s-s) scattering geometries contain distinct contributions from elasto-optic and ripple mechanisms. The elasto-optic contribution depends on \( \frac{\delta \varepsilon}{\varepsilon_0^{3/2}} \) (and hence on the elasto-optic tensor, \( k_{ijkl} \)) where \( \varepsilon_0 \) is the d.c. dielectric constant. The ripple contribution on the other hand depends only on the ripple amplitude at the film surface i.e. \( u_{z,q} (z = 0) \), where \( q_{\parallel} \) is the in-plane surface phonon wave-vector. In metals where \( \varepsilon_0 \) is often large, the elasto-optic contribution becomes negligible. On the other hand the EO contribution is significant in semiconductors where strong bulk modes are observed [25, 30]. Examples of excitations occurring in the polarized geometry are Rayleigh waves and generalized Lamb modes [18,25]. Stoneley modes that have recently been observed also fall in this category [22,33].

2. Depolarized (p-s and s-p) scattering geometries do not contain any contribution from ripple scattering. These modes are strong only in transparent media where the elasto-optic contribution is significant. Examples of depolarized scattering are Love modes [34,35].

Finally, figure 3.8 compares the theoretical scattering cross sections with experimental data from (110) GaAs with the in-plane wave-vector along [1 1 0]. The strong low energy peak corresponds to the Rayleigh mode that arise from ripple contributions. The weaker peaks appearing around 20 and 40 GHz correspond to the transverse and longitudinal bulk modes respectively. The intensities of these peaks depend upon combinations of different elastic and elasto-optic coefficients [28]. Figure 3.9 is a
similar spectrum from (001) GaAs showing a strong Rayleigh mode. The high energy shoulder alongside the Rayleigh mode is due to scattering from the Lamb wave continuum [25]. The bulk modes of figure 3.8 are absent in figure 3.9 because the combinations of elasto-optic coefficients on this surface gives rise to negligible scattering from bulk waves.
Figure 3.1: Normalized Rayleigh wave velocity on a free surface of an isotropic material (reference [9], pg. 49).

Figure 3.2: Sagittal plane displacement amplitudes for Rayleigh wave on a bulk isotropic unsupported layer. The amplitudes decay exponentially within a few wavelengths into the films (reference [9], pg. 50).
Figure 3.3: Co-ordinate system for wave propagation in thin layers (a) propagation direction sagittal plane; (b) sagittal plane co-ordinates for thin film and substrate (reference [9], pg. 38).
Figure 3.4: Rayleigh wave on an isotropic supported film. 1. Film stiffens substrate - only one Rayleigh mode present. 2. When the film loads the substrate, the number of supported modes increases with film thickness. Higher order modes labeled $R_2$ and $R_3$ are called Sezawa modes (reference 9, pg. 59).
Figure 3.5: (a) vertical ($u_3$) and (b) horizontal ($u_1$) displacement amplitudes for the third Rayleigh mode of figure 3.4.2 (reference [9], pg. 65).
Figure 3.6: Dispersion curves for W on Al (solid lines) and Al on W (dotted lines). The lowest order Rayleigh mode is asymptotic to the Stoneley mode in the former and the second order Rayleigh mode is asymptotic to the Stoneley mode in the latter (reference [9], pg. 75).

Figure 3.7: Sagittal plane displacement amplitudes for a Stoneley wave using material parameters of W and Al. Solid lines are W on Al and dotted lines refer to Al on W (reference [9], pg. 74).
Figure 3.8: Experimental and theoretical Brillouin cross sections for (110) GaAs with \( q \) along [110] (reference [28], pg. 1613).

Figure 3.9: Experimental and theoretical Brillouin cross sections for (001) GaAs with \( q \) along [100] (reference [28], pg. 1614).
References


CHAPTER IV

Elastic Properties of BCC Cobalt

The chapter presents the results of Brillouin light scattering off surface phonons in thin films of body centered cubic (bcc) cobalt films. The surface (Rayleigh) mode dispersion along various crystal directions on different crystal surfaces enables determination of the three elastic constants of this cubic material. The Rayleigh mode dispersion is analyzed using a model for an anisotropic supported film (chapter 3, appendix A).

1 Introduction

An understanding of the delicate interplay between configurational and magnetic energies in itinerant ferromagnets currently presents a critical test of first-principle calculations techniques to determine the electronic structure of metals [1-4]. This is especially challenging since the energy difference between two distinct crystal structures may be comparable to the small energy differences associated with magnetic effects. One such structure, cobalt, which can be stabilized as thin supported epitaxial ferromagnetic films in the naturally occurring hexagonal closed packed phase as well as face and body centered cubic configurations offers an excellent model system to test these calculations [5-9]. BCC cobalt also has the distinction of being one of the most strongly correlated 3d transition metals. Correlations may play an important
role in establishing the properties of the different phases of cobalt since the relative strength of the electron-electron interaction determines the competition between delocalization due to band formation and localization due to Coulomb coupling between electrons. Friedel et al have discussed the effects of correlations on the lattice constant and bulk moduli $B$ for 3$d$ elements and shown that for elements near the middle of the series, electron-electron interactions strongly influence these properties [2]. They also note that while non-magnetic mid-row 3$d$ elements should show substantial elastic softening compared with the zero correlation values, the softening of $B$ should be further enhanced when these elements become magnetic. A determination of the elastic properties of the different phases of cobalt will hence be valuable towards experimentally establishing the role of correlations on the bulk moduli of transition metals. The elastic constants hence not only provide direct insight into the microscopic nature of bonding, but also place important constraints on the reliability of elastic property calculations in magnetic materials. This project reports measurements of the three independent elastic constants of several relatively thick single crystalline Co films stabilized in the bcc phase. The elastic properties of bcc Co are determined from the Rayleigh phonon dispersion of the coupled layer (Co) - substrate (GaAs) system using Brillouin light scattering. The elastic constants determined via independent Rayleigh mode dispersions for propagation along different crystal directions on film surfaces of different orientations are in good agreement.
2 Sample Description

Recent advances in thin film growth techniques have enabled researchers to stabilize a number of artificial crystal structures up to a few 100 Å thick, by growing them on a suitably lattice matched substrate [5-10]. These structures are not found on a thermodynamic phase diagram. They are termed as metastable rather than forced structures because they are in epitaxial registry with the substrate that stabilizes their growth in a nearly strain free manner up to a few 100Å. The pioneering work on stabilizing bcc Co on GaAs by Prinz was motivated by a previous study of bcc Fe stabilized on a GaAs substrate [10], and lattice constant vs composition studies on Fe$_x$Co$_{1-x}$ alloys [11]. Since bcc Fe with a lattice constant $a_0 = 2.866 Å$ grows epitaxially on (110) GaAs, it was expected that bcc Co with a predicted lattice constant of 2.819Å should also grow on GaAs (figures 4.1 and 4.2).

Five bcc Co films grown on GaAs substrates by molecular beam epitaxy techniques described in [5,10] were utilized in this study. Two films 357Å and 202Å thick, were grown on a (110) GaAs substrate. The other three samples 106Å, 125Å and 216Å thick, were grown on the (001) face of GaAs. The epilayer thickness ($h$) was measured to an accuracy of about two monolayers. Each film was confirmed to be of bcc phase through X-Ray diffraction. The bcc structure of the two thickest Co films (357Å and 202Å) was also confirmed by conversion electron extended X-Ray absorption fine structure (EXAFS) [12]. BCC Co grows nearly strain free on GaAs with a substrate-lattice mismatch of < 0.25%.
3 Experiment

Figure 4.3 shows a schematic diagram of the experimental back-scattering geometry. Light with wave-vector \( k_i \) and angle of incidence \( \theta_i \) is incident on the sample and light with wave-vector \( k_s \) is inelastically scattered by surface and bulk excitations in the medium. Scattering can occur only from phonons that satisfy wave-vector conservation. The components of the phonon wave-vector parallel and perpendicular to the sample surface are

\[
q_\parallel = k_i \sin \theta_i + k_s \sin \theta_s \\
q_\perp = \eta (k_i \cos \theta_i + k_s \cos \theta_s)
\]

(4.1)

(4.2)

where \( \eta \) is the refractive index of the medium and the angles are as labeled in figure 4.3. In opaque media, the normal component of the photon wave-vector is severely attenuated by absorption (penetration depth \( \sim 120 \text{Å} \) in Co). Hence, the condition for \( q_\perp \) breaks down leading to a range of permissible \( k \) vectors for scattering and a consequent broadening of the spectrum. This broadening is severe in metals [13,14]. However, the surface wave-vector \( q_\parallel \) is still well defined by equation 4.1. Hence, scattering in metals occurs primarily off phonons traveling parallel to the surface. For back-scattering, \( \theta_l = -\theta_s \) and since \( k_i \approx k_s \),

\[
q_\parallel = 2k_i \sin \theta_i
\]

(4.3)

From energy conservation, we have

\[
\omega_s = \omega_i \pm \Omega
\]

(4.4)

where \( \Omega \) is the phonon frequency, \( \omega_i \) is the frequency of the incident laser light and \( \omega_s \) is the frequency of the scattered light. The \( \pm \) signs represent the anti-Stokes and Stokes processes that annihilate and create phonons respectively. Brillouin spectroscopy measures the frequency
shift $\Omega$ of the inelastically scattered light for a given in-plane wave-vector $q_y$. This dispersion relation is obtained by tuning the wave-vector $q_y$ through angle $\theta$.

Brillouin scattering measurements were performed at room temperature in a back-scattering geometry with a six pass tandem Fabry-Perot interferometer using the 5145Å Ar-ion and 6471Å Kr-ion laser line. Different sets of interferometer plates suitably coated for use with each of the two exciting laser lines were utilized. The incident beam was polarized in the sagittal plane (i.e. p-polarized) and the scattering was recorded in a p $\rightarrow$ p geometry when phonon features had to be isolated from spin wave contributions. Spectra were recorded using an incident laser power of 120-150 mW and a sampling time of 3 hours per spectrum. The in-plane wave-vector component ($q$) was tuned by varying $\theta$ between 30° and 70° (equation 4.3), measured to an accuracy of ±0.1°. The aperture was reduced from f/1.4 to f/3.5 at lower angles of incidence to compensate for increased broadening.

4 Results

Figures 4.4 and 4.5 show typical Brillouin spectra from the two thicker films [15]. Curves (1) and (2) of figure 4.4 were recorded at an angle of incidence, $\theta_i = 70°$ with $q$ along [001] and [110] respectively on the 357Å film. R identifies the Rayleigh mode while the shoulder labeled L arises from scattering by Lamb waves. Magnon scattering features have been suppressed in figure 4.4 through polarization selection. Figure 4.5 shows data from the 202Å film measured at $\theta_i = 60°$. In this case, spin wave features are retained by not analyzing the scattered beam.
Surface-like (S) and bulk-like (B0) magnons are observed in addition to the strong Rayleigh peaks (R). The magnetic nature of the latter excitations was confirmed through polarized (p → s) scattering and their response to a magnetic field [16]. Similar phonon features were observed in the (001) grown films.

The phase velocities, $v_R$, of the Rayleigh mode R were measured for propagation along the [001] and [110] directions in the (110) oriented films while velocities along [100] and [110] were determined in the (001) samples. In all cases $v_R$ was obtained by dividing the measured Brillouin frequency by the phonon wave-vector $q = 2k_0 \sin \theta$ where $k_0$ is the wave-vector of the incident beam. The measured phonon dispersions in the various samples are summarized in figures 4.6 and 4.7 where $v_R$ is plotted vs $q/h$. Data obtained using the 6471Å exciting laser line are indicated by filled circles while the hollow symbols indicate data acquired with 5145Å radiation. The solid lines in figure 4.6 and 4.7 are calculations (described below).

5 Discussion

In (110) grown bcc cobalt on GaAs, acoustic modes with $q$ along [110] and [001] consist of decoupled partial waves [17,18]. The principal sagittal plane surface mode is the Rayleigh wave. Solutions for the corresponding higher-order modes (Sezawa waves) exist only for cobalt film thickness' greater than those utilized in this study. The modes with displacements perpendicular to the sagittal plane are Love modes [17, 18]. These excitations are not observed in our measurements due to the lack of ripple contribution to these modes and the weakness of
elasto-optic scattering from opaque metal surfaces. The velocities of the Rayleigh waves are calculated by taking into account the mechanical boundary conditions at the film interfaces - vanishing of stress normal to layer at the free surface and continuity of displacement and stress across the interface.

The best fits to the measured Rayleigh velocity dispersion are calculated based on elastic continuum theory for a thin anisotropic film on a cubic substrate. The elastic constants of bulk hcp Co are used as initial inputs to the fitting procedure [19]. The bulk elastic parameters of the GaAs substrate are assumed to be unmodified due to epitaxial growth. The density of the epilayers is estimated \((8.83 \times 10^3 \text{ kg/m}^3)\) from the measured lattice constant of the 357Å thick bcc cobalt film [5]. The calculated dispersion for Rayleigh mode propagation along [001] and \([\bar{1} 1 0]\) in the two (110) oriented films are shown as solid lines in figures 4.6. The fits to the data for the two propagation directions were obtained with a single set of \(c_{11}, c_{12}\) and \(c_{44}\) constants. While the fits provide a good overall description, the data for the same \(qh\) (=0.43) from the two films lie slightly outside their error margins. This feature could reflect some differences between the elastic constants of the two films, although such differences can be accounted for within the quoted errors placed on the \(C_i\)'s. As discussed below strain effects may give rise to these small modifications. The second column in table 4.1 summarizes the resulting best-fit elastic constants for these two films. The \(c_{11}\) (193 GPa) and \(c_{44}\) (48 GPa) values for the (110) bcc cobalt films are considerably smaller (reductions of 40% and 35%
respectively) than those already reported for the bulk hcp phase. The \(c_{12}\) (163 GPa) constant is not changed significantly from the hcp value.

The elastic constants for the (001) oriented 106Å, 125Å and 216Å epilayers are similarly obtained by fitting the corresponding \(v_R\) for [100] propagation. The use of two different excitation wavelengths enables access over a wider range of \(q_h\) in these films of limited thickness. The results of the fits are shown as solid lines in figure 4.7 and yield the following values for the elastic constants; \(c_{11} = 212\) GPa, \(c_{12} = 165\) GPa and \(c_{44} = 53\) GPa. The error margins on these values are slightly larger than those deduced for the (110) films discussed above, and reflect in part the restrictions on the extent of \(q_h\). The elastic constants determined independently from the bcc Co films of different orientations are in good agreement with each other (table 4.1). The Rayleigh wave dispersion along the \([ \bar{1}10]\) direction in the 106Å (001) film was also determined as a further check of the consistency of the results. The inset in figure 4.7 illustrates these results. It shows excellent agreement between the measured Rayleigh velocities along the \([ \bar{1}10]\) direction for the 106Å film and the calculated \(v_R\) utilizing the \(C_{ij}\)'s determined from the dispersion for \(q\) parallel to [100].

In evaluating error margins associated with the elastic constants extracted from the Rayleigh wave dispersions, a threshold error \(T = \sum_{i=1}^{N} e_i\), where \(e_i\) is the error associated with a measurement of \(v_R\) was established for wave propagation along [001] and \([ \bar{1}10]\) in the (110) grown films. Those values of \(C_{ij}\)'s that satisfied \(\sum_{i=1}^{N} [v_{R_i}^{\text{exp}} - v_{R_i}^{\text{calc}}] < T\) (summation index \(i\)
over the total number of data points) simultaneously for propagation along both directions were considered acceptable. The errors $e_i$ varied between 0.5% and 2% with the largest contribution to $e_i$ being derived from the measurement of the Brillouin shift. The uncertainties assigned to the elastic constants represent the spread in acceptable values of these constants as demanded by the threshold criteria. These error margins on $C_{ij}$ can hence be viewed as a measure of the sensitivity of the Rayleigh velocity in the thin films to the corresponding elastic constants. Even though the Rayleigh wave is often most sensitive to $c_{44}$, in this case the error margins associated with the other elastic constants are comparable to that deduced for $c_{44}$. The reason for this most likely arises from the fact that $c_{11}$ in these films is about four times larger than $c_{44}$.

In a recent paper, Liu and Singh [20] theoretically calculated the elastic constants of *bulk* bcc cobalt. Their reported values for $c_{12}$ and $c_{44}$ are significantly higher than those measured in the supported epilayers in this study. However, there is good agreement between their calculated and our measured $c_{11}$. The results of [20] are included in table 4.1 along with the elastic constants of the hcp and fcc phases of Co (fcc Co not a thin film; obtained by rapid quenching from melt). It is noted in [20] that the calculated elastic constants, together with the minimal lattice mismatch between epilayer and GaAs substrate, are generally consistent with the epilayer maintaining coherency to thickness' of the order of tens of Å. Beyond this critical thickness the interfacial energy will be unable to stabilize the bcc phase. Given the reduced elastic constants for the bcc cobalt films measured in our experiments, it is conceivable that the
interfacial energy remains dominant and continues to stabilize the bcc phase in our films beyond the critical thickness of about 50Å [21]. In fact, the samples investigated in this work are amongst the thickest bcc phase Co films synthesized. It is worth noting that the calculated $C_{ij}$'s in [20] represent an unstable crystal configuration; i.e. ($c_{11} < c_{12}$) whereas our measured values for $c_{11}$ and $c_{12}$ though close in magnitude still represent a stable structure.

In another work Marcus et al calculated the elastic constants of bcc Co using a full potential linear tin muffin potential [22]. This method is believed to be more accurate than the linearized augmented plane wave (LAPW) method used in [20]. The value of $c_{12}$ from this calculation is not far removed from our $c_{12}$ from Brillouin measurements (table 4.1). However there is no agreement in the values of $c_{11}$ and $c_{44}$. At the same time, the theoretical values of $c_{44}$'s from [20] and [22] agree well. After comparing the results from various calculations and our measurements, it was suggested (Marcus) that the measured $c_{11}$ and $c_{12}$ values are correct but more work might be needed to verify the value of $c_{44}$ [23]. Hence a measurement of the $C_{ij}$'s of epitaxially stabilized fcc Co using BLS, would be a useful test.

BCC Co has also recently been stabilized on a non-cubic tetragonal TiAl substrate[24]. This work represents the first effort at stabilizing metastable bcc Co on a non-cubic substrate. Quantitative low energy electron diffraction (QLEED) was used to measure the normal and in-plane strains at the interface using the theoretical values of the elastic constants [22]. QLEED estimates of the bulk modulus of bcc Co gave $B = 2.54 \times 10^{12}$ N/m² which agrees with the
result from calculations [22] \((2.74 \times 10^{12} \text{ N/m}^2)\). It must be pointed however that the QLEED method is not as accurate as Brillouin light scattering [22,23].

The reasons for the reduction in the measured \(c_{44}\) in the bcc phase are yet unclear. One possible explanation lies in the nature of the Brillouin light scattering measurements. Though the experiment probes the surface phonon dispersion, these excitations decay well into the film. Thus bulk rather than surface elastic properties are being probed. Any inhomogeneities in the film or substrate by way of As inter-diffusion, presence of impurity contaminants like carbon [5], strain relaxation with thickness etc. will modify the Rayleigh wave dispersion. Since the Rayleigh mode dispersion is sensitive to \(c_{44}\), any growth related effects that modify the Rayleigh mode dispersion will influence \(c_{44}\).

Reduction in elastic constants can also arise from changes to the lattice parameters (strain effects); the measured lattice constant of the 357Å film \((a_0 = 2.827\text{Å})\) is about 3% larger than the value predicted in [20]. This underestimation in the calculated \(a_0\), while typical of local spin density approximations, is not adequate to explain the dramatic reduction in \(c_{44}\) that we have found. Strain effects can lead (through higher order terms) to changes in the elastic constants and to a thickness dependence of the \(C_{ij}\)'s. However, the close lattice match between epilayer and substrate which aided in the choice of GaAs as a substrate, would argue against the significance of such effects. The small differences in \(C_{ij}\)'s between the (110) and (001) layers may be due to differences in film quality associated with growth along distinct orientations.
Aside from extrinsic factors such as the growth rate, specific fabrication conditions and structural defects, strong electron-electron correlations that characterize the bcc Co films could also influence the elastic properties in relation to the hcp phase. This connection between correlations and elastic stability in transition metals was first discussed by Friedel [2]. The relative strength of the electron-electron interaction is governed by $U/W$, where $U$ and $W$ are the intra-atomic Coulomb energy and the band width respectively. The band width of metastable bcc cobalt is approximately 1.5 eV smaller than that of hcp Co [25], so that $U/W$ of bcc cobalt is ~30% larger than that of the thermodynamically stable hcp modification. Correlations were shown to reduce the bulk modulus $B$, the reduction being strongest near the middle of $3d$ series [2]. From table 4.1 the measured value of $B (= [c_{11} + 2c_{12}]/3 = 178$ GPa) for the bcc cobalt films is reduced by 10% from the hcp value [19]. Estimates based on equation 9 of reference [2] of the relative change between the bulk moduli in the hcp and bcc bulk phases of Co suggest that correlations would result in ~30% reduction in $B$. The fact that our Co epilayers are in registry with the substrate would however tend to inhibit effects of correlations on the lattice expansion that would otherwise occur in the bulk phase. This constraint on the in-plane lattice parameters may be important in accounting for the degree of suppression we have determined from hcp to bcc in $c_{11}$ and $c_{44}$. However, the effects of extrinsic causes discussed above must be systematically examined prior to identifying electron-electron correlations as an important reason for reduced $C_{ij}$'s measured in the bcc cobalt films.
6 Summary

This chapter discussed measurements of elastic constants of the bcc phase of Co by Brillouin light scattering techniques. The measured Rayleigh phonon dispersion curves of Co on GaAs are well described by elastic continuum theory. The $c_{11}$ and $c_{44}$ elastic constants extracted from the Rayleigh mode dispersions are reduced by almost 40% and 35% from their respective values in the thermodynamically stable hcp phase. $C_{ij}$'s determined independently from Rayleigh wave dispersions along distinct crystallographic directions in films of different orientations agree well. There is controversy between various calculations and experiment on the values of $C_{ij}$'s for bcc Co. While some agreement exists between theory and experiment on the values of $c_{11}$ and $c_{12}$, there is a large discrepancy between the calculated and measured $c_{44}$'s. Some possible reasons for these differences between experiment and theory have been outlined. The elastic constants of epitaxially stabilized thin films of fcc Co will be a useful measurement for comparison with theory. Additional theoretical insight into the role of electron correlations on the elastic properties of this strongly correlated 3$d$ metal would also be valuable.
Figure 4.1: Unit cell and atomic arrangement of (1 TO) face of GaAs and bcc Fe lattice grown on GaAs (reference [10]).

Figure 4.2: Lattice constant vs composition for Fe-Co system. The close lattice match between bcc Co and GaAs makes the latter a natural choice for substrate (reference [5]).
Figure 4.3: Back-scattering geometry of Brillouin experiment. The wave-vector component parallel to the surface ($K_R$) scatters light strongly in metals (reference [14]).
Figure 4.4: Brillouin spectra from the (110) grown 357Å bcc Co film in p → p scattering geometry and angle of incidence $\theta_i = 70^\circ$. 1. In-plane wave-vector (q) parallel to [001]; 2. q parallel to [110]. R identifies Rayleigh mode and L the Lamb wave continuum.
Figure 4.5: Brillouin spectra from the (110) grown 202Å bcc cobalt film at $\theta_1 = 60^\circ$. The scattered radiation was not analyzed. R identifies Rayleigh mode while $S$ and $B_0$ are surface-like and bulk-like magnons 1. $q$ parallel to [001]; 2. $q$ parallel to [$\bar{1}10$].
Figure 4.6: Rayleigh wave dispersion data together with calculation (solid line) for the two (110) grown films. Hollow symbols are data taken at an incident wavelength of 5145Å while the solid symbols refer to data taken at 6471Å. 1. Dispersion for q parallel to [001]; 2. q parallel to [110].
Figure 4.7: Rayleigh wave dispersion data plus calculation (solid line) for the two (001) grown films for $q$ parallel to [100]. Hollow symbols are data taken at an incident wavelength of 5145Å while the solid symbols refer to data taken at 6471Å. The inset shows measured Rayleigh velocities along [110] for the 106Å film. The solid line is the calculated velocity utilizing elastic constants derived from data for in-plane wavevector transfer parallel to [100].
Table 4.1: Elastic constants of bcc Co \((x10^9 \text{ N/m}^2)\).

<table>
<thead>
<tr>
<th>(C_{ij})</th>
<th>BCC Co measured (^a)</th>
<th>BCC Co measured (^b)</th>
<th>BCC Co calculated (^c)</th>
<th>BCC Co calculated (^d)</th>
<th>HCP Co measured (^e)</th>
<th>FCC Co (^f)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(c_{11})</td>
<td>193 ± 10</td>
<td>212 ± 21</td>
<td>205</td>
<td>398</td>
<td>307</td>
<td>242</td>
</tr>
<tr>
<td>(c_{12})</td>
<td>170 ± 12</td>
<td>165 ± 17</td>
<td>278</td>
<td>212</td>
<td>165</td>
<td>160</td>
</tr>
<tr>
<td>(c_{44})</td>
<td>48 ± 6</td>
<td>53 ± 10</td>
<td>152</td>
<td>143</td>
<td>75</td>
<td>128</td>
</tr>
<tr>
<td>(B)</td>
<td>178 ± 12</td>
<td>181 ± 18</td>
<td>254</td>
<td>274</td>
<td>193</td>
<td>187</td>
</tr>
<tr>
<td>(G)</td>
<td>12 ± 7</td>
<td>24 ± 19</td>
<td>&lt;0</td>
<td>93</td>
<td>71</td>
<td>41</td>
</tr>
</tbody>
</table>

Measured elastic constants for bcc Co in (GPa) derived from fits to the Rayleigh wave dispersion data. Calculated and measured elastic constants for various phases of Co are included for comparison. \(B\) is the bulk modulus \(B = \frac{1}{3} \left( \frac{c_{11} + 2c_{12}}{3} \right) \) and \(G = \frac{c_{11} - c_{12}}{2} \).

- \(^a\) This work - 357Å and 200Å (110) grown films on GaAs.
- \(^b\) This work - 106Å and 125Å (001) grown films on GaAs.
- \(^c\) Reference [20].
- \(^d\) Reference [22].
References


[23] Private communication from P.M. Marcus.


CHAPTER V

Magnons in Ferromagnetic Films

This chapter introduces the theory of spin waves propagating in the bulk and on surfaces of ferromagnetic films. BLS is a powerful tool for probing spin wave dispersion and yields valuable information on magnetic properties like the exchange constant and anisotropies.

1 Introduction

Magnons or spin waves are used to describe small perturbations in the ordering of magnetic moments in a lattice. Like all quasi-particle excitations, magnons possess energy and momentum which are related by the theory of spin waves. Materials where the atomic moments are aligned parallel are called ferromagnets. A local moment Heisenberg hamiltonian provides an adequate description of insulating ferromagnets whereas ferromagnetism in 3-d transition metals (Fe, Co, Ni) requires a complex itinerant model for a complete understanding.

Many salient features of spin waves in 3-d metals studied using BLS can be illustrated using a simple local moment picture. The quantum mechanical description of the system begins by writing a hamiltonian in terms of electron spins that are responsible for ferromagnetic ordering [1,2]. The hamiltonian with a magnetic field $H$ along the $z$ axis has the form

$$ H = -J \sum_{i,\delta} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta} - g \mu_B H \sum_i \mathbf{S}_i $$  \hspace{1cm} (5.1)
where $J$ is the Heisenberg exchange integral, $g$ is the spectroscopic splitting (Lande) factor and $\mu_0$ is the Bohr magneton. The vector $\delta$ connects a spin at site $i$ to its nearest neighbors. The microscopic spin wave theory is derived using the second quantization method devised by Holstein and Primakoff [3,4], where the spins $S_i$'s are replaced by magnon creation/annihilation operators. The resulting magnon dispersion relation is given by

$$E_q = \hbar \nu_q = (J_0 - J_q) + g \mu_B H \quad \text{where} \quad J_q = 2SJ \sum_{\delta} \exp(iq\delta)$$

(5.2)

In the long wavelength limit $|q\delta| \ll 1$. Thus

$$J_0 - J_q = JS \sum_{\delta} (q\delta)^2$$

(5.3)

In cubic materials with lattice constants $a$, we obtain

$$\omega_q = \gamma H + \left( \frac{2JS}{\hbar} \right)(qa)^2 = \gamma (H + Dq^2)$$

(5.4)

where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio and $D = 2JSa^2/g\mu_B$ is the exchange stiffness constant. For $g = 2.0$, the gyromagnetic ratio is 3.8 GHz/kOe. The exchange parameter $D$ in most ferromagnetic materials ranges from $(1-5) \times 10^9$ Oe-cm$^2$. Hence exchange field $(Dq^2)$ contributions range from a negligible amount near $q = 0$ to several kOe for $q \sim 10^6$ cm$^{-2}$. The long wavelength approximation requires $q \ll \pi a$ [5].

Figure 5.1 shows the precession of spins in a chain. Each spin precesses on a cone and the motion is described by the oscillation of its transverse component $S^x_i = iS^y_i = S^0_i \exp[i(\omega_q t - qr_i)]$ and the phase difference between adjacent spins is given by
\[ \Delta \varphi = qa = \frac{2\pi a}{\lambda} \] The exchange interaction discussed above is a short range interaction and its strength falls exponentially with separation.

The magnetic dipole-dipole interaction is a long range interaction and contributes significantly to the energy of long wavelength magnons. The magnetic dipolar contribution is evaluated by calculating the dipolar field \( h(t) \) created by oscillations in the transverse magnetization \( m(t) \) of the spin wave. In a semi-classical treatment the oscillations in \( m(t) \) and \( h(t) \) (\( m(t) \perp M \parallel H \)) are obtained by solving Maxwell's equations with appropriate boundary conditions, where \( M \) is the saturation magnetization [6]. If \( \theta_k \) denotes the angle between the saturation magnetization and the wave-vector \( \mathbf{q} \), the bulk ferromagnet magnon dispersion relation (equation 5.4) takes the form [3]

\[
\omega_q^2 = \gamma^2 (H + Dq^2)(H + Dq^2 + 4\pi M\sin^2 \theta_k) \quad (5.5)
\]

The spin wave dispersion of equation 5.5 is illustrated in figure 5.2. The lower branch for \( \mathbf{q} \parallel M \) extrapolates to the Zeeman frequency \( \omega_\parallel = \gamma H \) in the limit \( q = 0 \). The gyroscopic precession cone for this branch is circular in the absence of dipolar contributions. When spin waves propagate away from \( M \) there is a finite dipolar contribution which becomes maximum when \( \mathbf{q} \perp M \). This leads to an increase in the magnon frequency and an elliptical precession of the spin waves. The dipolar interaction broadens the magnon spectrum (spin wave manifold) as indicated by the hatched lines in figure 5.2. Before the advent of Brillouin spectroscopy, the spin wave dispersion (equation 5.5) could only be verified using ferromagnetic resonance
(FMR). This is a uniform precession of spin waves corresponding to the $q = 0$ case \cite{7,8}. We note that while exchange fields are significant only at large $q$'s, dipolar fields ($4\pi M$) that are of the order of 10-20 kGauss in ferromagnetic metals are always significant. When exchange is neglected the spin waves are called magnetostatic modes.

In addition to exchange and dipolar fields there exist anisotropy fields such as magnetocrystalline fields that arise from spin-orbit coupling. Magnetocrystalline fields represent variations in the crystal magnetic field along different directions and possess the same symmetry properties as the crystal. There also exist surface and volume anisotropies that arise from interfaces and their properties are strongly influenced by growth conditions such as substrate temperature, growth rate and lattice mismatch. The origins of these anisotropies are difficult to quantify and most work on interface related anisotropies has been conducted using phenomenological models. We show in chapter VI that uniaxial volume anisotropies in bcc Co arise from magnetoelastic effects driven by interface strains. The magnitudes and symmetries of various anisotropies determines the overall symmetry of the in-plane magnon dispersion. The subject of crystal anisotropies is discussed in chapter VI and neglected below for simplicity.

2 Modes of a Thin Slab

While solving the problem of magnetostatic spin waves in an infinite, isotropic ferromagnetic slab, Damon and Eshbach found a class of solutions corresponding to surface spin waves \cite{9}. A solution of the magnetic equation of motion (neglecting exchange) and Maxwell's equations
gives rise to a set of dispersion curves shown in figure 5.3. The parameters labeled in figure 5.3
are given by the following expressions

\[ \Omega_B = [(\Omega_H (\Omega_H + 1))]^{1/2}, \quad \Omega_S = \Omega_H + 1/2 \quad \text{where} \quad \Omega_H = \frac{H}{4\pi M} \]  

(5.6)

The geometry of the calculation corresponds to a slab in the \( yz \) plane with \( H, M \) along \( z \) and the
film normal along \( x \). No restrictions are imposed on the in-plane wave-vector components \( q_y \)
and \( q_z \) and the solutions are a continuum of traveling magnetostatic waves. The wave-vector
component normal to the slab surface \( (q_x) \) can be real or imaginary corresponding to bulk-like
and surface-like waves respectively. The bulk waves obey a dispersion law similar to equation
5.5 with \( D = 0 \). Solutions with imaginary \( q_x \) describe waves localized to the surface whose
amplitudes decay exponentially into the slab.

The series of sheets in figure 5.3 between \( \Omega_1 \) and \( \Omega_0 \) represent the bulk manifold.
Figure 5.4 shows the family of volume branches for \( q = q_z \) (parallel to \( M \)) along with their field
distributions across the film. Each sheet corresponds to a standing wave solution with \( q_x =
(n\pi\ell/h) \) where \( h \) is the slab thickness. The \( n = 0 \) mode is the constant FMR mode. As the wave-
vector moves away from the magnetization axis \( (z) \) the bulk manifold collapses and a new sheet
emerges at a critical angle \( \Theta_c \) which corresponds to the Damon-Eshbach (DE) mode or the
surface mode [9]. The frequency of this mode increases until \( q \) is directed along \( y \) (i.e. \( \perp M \)).
The mode dispersion along this direction is given by

\[ \Omega^2 = \Omega_B^2 + [2(1 + \coth(|q_y|))]^{-1} \]  

(5.7)
while the bulk manifold has a single degenerate frequency $\Omega_{0}$. Figure 5.5 shows the surface wave dispersion for $q \perp M$ along with its exponentially decaying amplitude profile. When $q_{y} = 0$, the surface mode is degenerate with the bulk mode and has a frequency $\Omega_{0}$. Its dispersion is linear for $q_{y}h \ll 1$ and saturates to a constant $\Omega_{0}$ for large $q_{y}h$.

When the surface wave propagates perpendicular to the magnetic field (i.e. $q = (q_{x}, q_{y}, q_{z} = 0)$) we obtain imaginary solutions for $q_{x} = \pm iq_{y}$. This gives rise to exponentially decaying or growing amplitudes i.e. $\exp(\pm q_{y}h)$ as $h \to \infty$, depending upon the sign of $q_{y}$. Thus, the free surfaces of the slab permits localization of spin waves. The upper and lower surfaces each support a localized spin wave propagating along opposite directions. Hence surface magnons exhibit a non-reciprocal or "one-sided" behavior (figure 5.6). The surface wave appears on either the Stokes side or the anti-Stokes side of the spectrum, but not on both. The larger the value of $|q_{y}|$ the more the mode gets localized at the surface and it decays in amplitude with a characteristic length of $1/|q_{y}|$. The propagation direction of the surface mode is defined by the vector $(\hat{B} \times \hat{n})$ where $\hat{n}$ is a unit vector directed away from the film surface. Hence if a spin wave propagates from left to right on the top surface of the slab, it will propagate from right to left on the bottom surface. If the surface wave appears on the anti-Stokes side, a reversal of $H$ will cause it to switch to the Stokes side and vice versa. The same switching can be effected by changing the sign of the in-plane wave-vector $\mathbf{q}$ (by rotating the incident field about the surface by $180^\circ$, figure 5.6) [10]. When the propagation direction of the DE mode approaches the critical angle $\Theta_{c}$ it loses its non-reciprocal nature.
and becomes a bulk-like reciprocal mode, appearing on both the Stokes and anti-Stokes sides of the spectrum. The angle $\Theta_S$ is determined from the relation $\tan \Theta_S = \Omega_{i f}^{1/2}$.

When exchange is included the degeneracy of the volume branch at $\Omega_{h}$ for $q_z = 0$ is lifted [11-13]. The magnon frequency is raised by the presence of the exchange term $Dq^2$. The surface branch sheet in the DE theory surface sheet now acts as a transition between adjacent volume sheets in the space $(\Omega, q_x, q_y)$. Figure 5.7 shows the removal of the degeneracy in the volume mode by including exchange and the intersection of the surface branch with a series of bulk modes. The modes repel each other at the crossing points [11]. When the surface sheet intersects with a volume branch there is strong mode mixing and the surface wave loses its non-reciprocal character. In a thin slab, the discrete bulk modes are well separated due the exchange term $Dq_x^2 (q_x = n\pi h)$. As the slab thickness increases, the exchange modes become closely spaced and form a continuum of states and the dispersion relation is now given by 5.5.

3 Light Scattering from Magnons

Initial theories on magnon light scattering examined the magnetic dipole interaction as a possible governing mechanism, where magnon creation or annihilation is accompanied by a unit change in the magnetic quantum number [14,15]. This mechanism however gave rise to a very small scattering cross section. Elliott and Loudon suggested that magnon scattering takes place due to an electric dipole transition where the magnetic quantum number changes due to a spin orbit interaction at the excited level [16,17]. The scattering cross section obtained here was
several orders of magnitude larger than before. Currently, the spin orbit mechanism is believed to be the dominant contributing factor to magnon scattering.

The magnon scattering cross section is evaluated by expanding the dielectric tensor in terms of spin operators [18-23]. The interaction Hamiltonian is given by

$$H_{\text{int}} = \sum_{\alpha, \beta, R} E_{\alpha}^{\alpha} \Delta \epsilon_{\alpha \beta} (R) E_{\beta}^{\beta}$$

(5.8)

where the subscripts $i$ and $s$ refer to the incident and scattered radiation and $R$ is a co-ordinate point. The dielectric tensor is expanded in terms of the magnetization vector as

$$\Delta \epsilon_{ij} = f_{ijkl} M_k + g_{ijkl} M_k M_l$$

(5.9)

The elements of tensors $f$ and $g$ are complex quantities when dissipation is included. The first term ($f$) gives rise to magnetic circular birefringence (Faraday rotation, $\phi_{MCB}$). The second term ($g$) corresponds to magnetic linear birefringence (Cotton-Mouton effect, $\lambda_{MLB}$). One magnon scattering terms in $\Delta \epsilon$ are linear in the transverse magnetization $M^+ = \mathcal{M}^X \pm i \mathcal{M}^Y$. Contributions from $f$ may be expressed as [18,20]

$$H_{\text{int}}^f = F \sum_{R} [(E_{i}^{Z} E_{s}^{+} - E_{i}^{+} E_{s}^{Z}) \mathcal{M}_{R} - (E_{i}^{Z} E_{s}^{-} - E_{i}^{-} E_{s}^{Z}) \mathcal{M}_{R}^{+}]$$

(5.10-a)

Similarly, the one magnon contributions from $g$ that is quadratic in $M_i$ takes the form

$$H_{\text{int}}^g = G \sum_{R} [(E_{i}^{Z} E_{s}^{+} + E_{i}^{+} E_{s}^{Z})(\mathcal{M}_{R}^{+} \mathcal{M}_{R}^{Z} + \mathcal{M}_{R}^{Z} \mathcal{M}_{R}^{+}) + (E_{i}^{Z} E_{s}^{-} + E_{i}^{-} E_{s}^{Z})(\mathcal{M}_{R}^{+} \mathcal{M}_{R}^{Z} + \mathcal{M}_{R}^{Z} \mathcal{M}_{R}^{+})]$$

(5.10-b)

$\mathcal{M}_{R}^{Z}$ refers to the magnetization direction at point $R$ lying in the film plane. The factors $F$ and $G$ depend upon the symmetry properties of $f$ and $g$. In simple cubic crystals most elements of $f$
and $g$ vanish from symmetry arguments [23]. The interaction hamiltonian contains information regarding the dependence of the scattered light intensity on the polarization ($\Delta \delta$) of the incident light, crystal symmetry and magnetization direction. The form of the hamiltonian provides the polarization selection rule for magnon scattering; i.e. light scattered from magnons is rotated 90° relative to the polarization of the incident light.

The magnetization vectors can be expanded in terms of magnon creation and annihilation operators [3], and the hamiltonian evaluated using Greens function techniques to obtain the scattering cross section [24]

$$\left( \frac{d^2 \sigma}{d\Omega d\omega} \right) \sim \frac{\hbar \gamma_0 \omega^2 c q^4 M}{\epsilon_0} \left\{ \frac{(N(\omega) + 1) |A^-|^2}{N(\omega)|A^+|^2} \right\}$$

where ± corresponds to the anti-Stokes and Stokes cross sections respectively and $N(\omega)$ is the Bose thermal occupancy factor. The $A$'s now contain all the information in $\Delta \delta$ (or $F, G$). Expressions for $A^-$ and $A^+$ are derived in [23] for spin waves in cubic crystals.

We now discuss various reasons for the large observed differences in the Stokes/anti-Stokes (S/aS) scattering intensities in different materials. From equation (5.11) above we note that thermal population effects could be one possible cause. However a simple calculation of the S/aS ratio ($= \exp(h\omega / k_B T)$) at room temperature rules out thermal effects as a possible reason. Wettling et al [23] have shown that interference effects between circular ($\phi_{\text{circ}}$) and linear birefringence ($\psi_{\text{lin}}$) terms (i.e. differences in the magnitudes of $A^-$ and $A^+$) explain the observed S/aS asymmetries in transparent materials like YIG [21,23], EuO [14], and other
transparent materials where the optical penetration depth is large. The scattered power when separated into $\phi_{MCD}$ and $\Psi_{MLB}$ can be expressed as

$$\left(\frac{P_{sc}}{P_{inc}}\right) \sim \left[ (\Psi_{MLB} \mp \Phi_{MCD})^2 + \left(\frac{1}{2}\Psi_{MLB} \pm \frac{1}{4}\Phi_{MCD}\right)^2 \right]$$

(5.12)

where the $\pm$ terms refer to the Stokes and anti-Stokes processes respectively. Note that the above two mechanisms are independent of magnetic field reversal.

Recent experiments have showed that large S/aS asymmetries persist even when interference effects are absent (ex. thick films of Fe) [10,25,26]. The reason for this asymmetry is that the eigenvectors of the created and annihilated waves are not equivalent. This inequivalence has been viewed as a localization effect due to the presence of a surface. Thus a surface spin wave propagating at the top surface of a slab (geometry of figure 5.3) must have its largest amplitude near the top surface while a surface spin wave propagating in the opposite direction must be localized at the bottom of the slab. If the scattering geometry is such that a spin wave is created at the top and destroyed from the bottom, then in strongly absorbing materials the incident light 'sees' more of the spin wave amplitude near the top surface than at the bottom surface. Hence we expect a large Stokes intensity and a weak anti-Stokes intensity.

In very thin opaque films the above argument for scattering intensity asymmetries is invalid. When $h \sim 200\text{Å} (<\text{amplitude decay length } 1/q_p)$ the amplitude of a magnon propagating localized at the lower surface is hardly attenuated when it reaches the top and vice-versa. Hence the incident light should perceive almost identical amplitudes from magnons
localized at both interfaces and give rise to similar S/aS scattering intensities. However a large S/aS asymmetry still persists in this case. Its origin can be traced to off diagonal contributions from the spin-spin correlation functions \( <S_s S_s> \), which are defined in [27] as

\[
S_y(x, x', t - t') = < S_i(x, t) S_j(x, t') >
\] (5.13)

Translational invariance allows us to express \( S_y(x, x', t - t') \) in terms of its Fourier transform and we obtain the correlation function in \((k, \omega)\) space. We follow the geometry of figure 5.3 where a slab is bounded by the planes \( x = (0, L) \) and extends infinitely in the \((y, z)\) direction. The diagonal element \( S_{yy}(q_y, \omega, x, x) \) is a measure of the square of the amplitude to find a spin fluctuation in the \( y \) direction, located at a distance \( x \) from the surface, with wave-vector \( q_y \) and frequency \( \omega \). The intensity of the scattered field in terms of the spin-spin correlation is given by

\[
I(q_y, q_z, \omega) = \frac{1}{2} J dx dx' \exp[i(\Delta q_y x - \Delta q_z x')] x[r_{yy}S_{yy} + r_{xx}S_{xx} + r_{yx}S_{yx} + r_{xy}S_{xy}]
\] (5.14)

The \( S_y \)'s are functions of \( q_y \) and \( \omega \) and the \( r_y \)'s are functions of the scattering geometry, refractive index, polarization etc. [25-27]. The perpendicular components of \( q_y \) are imaginary when the material is strongly absorbing.

The intensity difference between the Stokes and anti-Stokes peaks results from the change of \(+\omega\) to \(-\omega\) in the \( S_y \) functions [27]. The diagonal functions \( S_{xx} \) and \( S_{yy} \) are not symmetric under time reversal. However, time reversal followed by a reflection about the center of the slab at \( x = L/2 \) is a symmetry operation. In the case of \( S_y \) or \( S_{xx} \), application of time reversal changes the signs of \( S_x \) and \( S_y \) because of the axial nature of \( S \). When we reflect about the center of the slab \( S_y \) changes sign to \(-S_y\) but \( S_x \) does not because \( S \) is an axial vector.
Thus when \( \omega \) changes to \(-\omega\) the \( S_x \) functions remain unchanged whereas the \( S_y \)'s undergo a change in sign. If \( S_{xy} \) contributes positively to the Stokes scattering intensity then \( S_{yx} \) will have a negative contribution on the anti-Stokes side. Unlike the case of scattering from thick films where the amplitude of a spin wave localized at one interface, is considerably diminished at the other interface thus giving rise to a S/aS asymmetry, the above result does not depend upon the explicit nature of the coupling (i.e. dipole-dipole) or even the presence of a surface. Scattering from bulk waves will show the same asymmetry if the spins are slightly deviated from the center of the slab at \( x = L/2 \).

We note that the factors \( r_{xx} \) and \( r_{yy} \) are real and positive while \( r_{xy}, r_{yx} \) are complex \([25,26]\). The correlation functions \( S_{xx} \) and \( S_{yy} \) are real and positive whereas the off-diagonal elements \( S_{xy} \) are purely imaginary. \( S_{yx} \) is purely imaginary because the spins \( S_x \) and \( S_y \) are 90° out of phase. Since the overall scattering intensity is positive, \( S_{xy} \) contributes to scattering only when \( r_{xy} \) has an imaginary component. This happens only when the wave-vector of the incident light is complex, a condition satisfied only in the presence of strong optical absorption \([25,26]\). Hence, this mechanism for the S/aS asymmetries does not prevail in transparent media like garnets where absorption is weak \([23]\). The mechanism discussed above for asymmetries in the S/aS scattering intensities from very thin ferromagnetic films applies towards understanding the origins of S/aS asymmetries in light scattering studies from spin waves in bcc Co.
Figure 5.1: Schematic diagram of spin wave motion (reference [4], pg. 357).

Figure 5.2: Spin wave dispersion of equation (5.5) lead to a band formation (reference [5], pg. 255).
Figure 5.3: Dispersion of magnetostatic modes. At angle $\Theta_s$ a new sheet emerges from the bulk manifold which has a surface character. This is the Damon Eshbach mode (reference [5], pg. 258).

Figure 5.4: Family of volume branches for $q = q_v$ (parallel to $M$) along with field distributions across the film. Each sheet corresponds to a standing wave solution with $q_v = (n\pi h)$ (reference [5], pg. 259).
Figure 5.5: Surface spin wave dispersion for $q \perp M$ along with its exponentially decaying amplitude profile. When $q_y = 0$, the surface mode is degenerate with the bulk mode (reference [5], pg. 260).

Figure 5.6: Non-reciprocal nature of surface spin wave. The mode appears on either the Stokes or anti-Stokes side of the spectrum depending on the direction of the field and the polarization of the incident radiation (reference [10]).
Figure 5.7: 1. Removal of the degeneracy in the volume mode by including exchange and the intersection of the surface branch with a series of bulk modes. 2. Repulsion of modes at crossing points [11].
References


CHAPTER VI

Magnetoelastic Effects in BCC Cobalt

The chapter investigates the origin of magnetic anisotropies in body centered cubic cobalt films using Brillouin light scattering. The in-plane directional and wave-vector dependence of surface and bulk magnon frequencies in (110) and (001) oriented films indicate a two-fold anisotropy in these cubic ferromagnets with the easy axis of magnetization lying in-plane along [001] and [100] respectively. Using elastic constants determined from phonon dispersion measurements, the origins of the observed uniaxial anisotropy is attributed to magnetoelastic interactions driven by lattice mismatch at the film-substrate interface. The easy axis in the (110) film is evident by the transformation of the surface spin wave into the $n = 0$ bulk magnon when the wave-vector lies parallel to the easy axis of magnetization.

1 Introduction

Advances in thin film growth techniques have enabled new phases of magnetic materials to be stabilized. One such material, cobalt, that naturally occurs in the hexagonal closed packed phase, has been grown in stable fcc and bcc phases as thin supported epitaxial films [1-3]. Studies on phases of Co have concentrated on understanding the observed tendency of the easy magnetization axis to display a thickness and orientational dependence of the magnetoelastic...
interactions and strain fields induced by interface lattice mismatch [5-10].

Despite the success of some of these models the specific origins of magnetic anisotropies in thin films are not fully understood. The problem is that the magnetic properties of thin films are very sensitive to the microscopic structure of the interface. Growth conditions, interface strains, roughness and inter-diffusion are all factors which determine magnetic anisotropies. The importance of the interface has been observed for example in thin (< 50Å) films of epitaxial bcc Fe on GaAs, where the roles of the easy and hard magnetization axes reverse at reduced film thickness due to interface anisotropies [11]. Similar effects have also been observed in Fe films (< 100Å thick) on tungsten substrates [12,13].

This chapter examines the character and origin of magnetic anisotropy in the bcc phase of cobalt using Brillouin light scattering from propagating surface and bulk spin wave excitations. Two bcc Co films of (110) and (001) orientations were stabilized on GaAs substrates as thin films with thickness of the order of 200Å. The two lowest energy branches of the spin wave manifold were observed and an analysis of their dispersion allowed determination of the anisotropy constants and exchange stiffness constants. Bulk anisotropies are clearly evident in the dispersion of the surface and bulk magnons in films of both orientations. In particular, a large in-plane uniaxial anisotropy contribution is required to account for the behavior of the magnon frequency as a function of the in-plane angle in the (110) film. It is shown, using elastic constants measured on the same bcc Co films, that
magnetoelastic effects arising from the small lattice mismatch strain largely account for this two-fold anisotropy.

2 Experimental Details

Two bcc cobalt films were grown by molecular beam epitaxy techniques as described elsewhere [3]. The films were 202Å and 216Å thick and oriented with the (110) and (001) planes parallel to their respective surfaces. The films were stabilized on a GaAs substrate that has a lattice mismatch of about 0.25% with the predicted lattice constant of bcc cobalt [3]. The epilayer structure was confirmed to be of the bcc phase through X-Ray diffraction and surface enhanced X-Ray absorption fine structure studies (EXAFS) [14]. The Brillouin scattering experiments were performed at room temperature in back scattering using a six-pass tandem Fabry-Perot interferometer. Single mode p-polarized light (150 mW power) at 5145Å and 6471Å was focused onto the sample with a typical sampling time of 2 hours. Spin wave excitations were mostly analyzed in the depolarized configuration in order to suppress phonon signals. The direction of $\mathbf{q}$ was systematically varied to lie along various in-plane directions. The in-plane propagation direction ($\phi_1$ defined below) was measured within an error of ±3°. A magnetic field was applied parallel to the layer plane and spin wave frequencies were measured for $\mathbf{q}$ propagating perpendicular to the applied field.
3 BLS Results

The dominant contribution to the lowest frequency spin waves in ferromagnets are anisotropies, dipolar magnetic fields and the external applied field. Frequencies of the surface and bulk modes can be calculated for thin ferromagnetic films including exchange interactions in a continuum limit that is valid for long wavelength spin wave excitations. The theory of dipole-exchange behavior begins by solving the equation of motion for the time and spatially varying magnetization, \( m(t) \), together with Maxwell’s equations of magnetostatics [15-20].

\[
\nabla \times h(t) = 0
\]

(6.1)

\[
\nabla \cdot (h(t) + 4\pi m(t)) = 0
\]

(6.2)

\[
\frac{1}{\gamma} \frac{\partial M}{\partial t} = M \times \left[ H - \frac{1}{M} \nabla_M E_{\text{ani}} + \left( \frac{2A}{M} \right) \nabla^2 M \right]
\]

(6.3)

where \( \gamma \) is the gyromagnetic ratio, \( h(t) \) is the dipolar magnetic field, \( M = M_0 + m(t) \) and \( H = H_0 + h(t) \). \( M_0 \) and \( H_0 \) are the static magnetization and the external magnetic field respectively. Both \( m(t) \) and \( h(t) \) satisfy the inequalities \( |m| \ll M_0 \) and \( |h| \ll H_0 \). In addition to the external and dipolar fields, there are effective field contributions from the exchange \( (\nabla^2 M) \) term and crystal anisotropies \( (E_{\text{ani}}) \). The gradient \( (\nabla_M) \) of \( E_{\text{ani}} \) is taken with respect to the direction cosines of the magnetization vector relative to the crystal axis. The crystal anisotropy fields contain contributions from four-fold and uniaxial anisotropies. The analysis gets complicated because the anisotropic fields depend upon the crystal orientation relative to the external magnetic field.
Therefore the magnetization direction that lies in the plane of the films does not align with the applied field for some geometries, due to the anisotropies present in the films.

The films are defined in the co-ordinate frame \((x', y', z')\) where the \(y'\) axis lies normal to the film planes and the \(z'\) axis lies along the \([001]\) \(([100])\) for the \((110)\) \((001)\) films. The spin wave calculations are performed in a co-ordinate system \((x, y, z)\) where \(y = y'\) and the \(z\) axis is oriented along the direction of saturation magnetization. The angle \(\phi\) is defined as the equilibrium angle between the \([001]\) \(([100])\) and the \(z\) axis for the \((110)\) \((001)\) films. A simple rotation about \(y'\) through angle \(\phi\) is used to transform from the primed to the unprimed reference frame (figure 6.1). \(\phi\) is calculated independently for each instance of crystal orientation by numerically minimizing the total free energy \(E\) with respect to \(\phi\). Working in the unprimed co-ordinate system reduces the number of equations of motion for \(\frac{dm}{dt}\) from three to two. We define \(\phi_1\) as the in-plane angle between the external magnetic field \(H_0\) and the crystallographic \([001]\) direction for the \((110)\) film, and the \([100]\) direction for the \((001)\) film. \(\phi_1\) is used to reference the propagation direction of the in-plane magnon wave-vector \(\mathbf{q}\) which is always directed perpendicular to \(H_0\). Note that the effective angle between \(H_0\) and \(M_0\) in the rotated co-ordinate frame is \((\phi_1 - \phi)\).

Since a calculation of the equilibrium angle requires \(E\) as a function of \(\phi\) while the equations of motion require the free energy in terms of the magnetization direction cosines, it
is instructive to write expressions for $E$ in both forms. The films are magnetized in-plane and hence all out-of-plane terms ($m'_y, K_P$) can be neglected while calculating $\phi$.

$$E^{(110)} = -H_0 M_o \cos(\phi_H - \phi) - K_U \cos^2 \phi + \frac{K_1}{4} \left(4\sin^2\phi - 3\sin^4\phi\right)$$

$$E^{(001)} = -H_0 M_o \cos(\phi_H - \phi) - K_U \cos^2 \phi + \frac{K_1}{4} \left(\sin^2 2\phi\right) \tag{6.4}$$

The form of $E$ in the primed frame expressed as magnetization direction cosines, in terms of the Zeeman, demagnetization, magneto-crystalline and uniaxial energies contributions is given by [21,22]

$$E^{(110)} = -H.M\frac{K_U}{M^2}m_z^2 + \frac{K_P}{M^2}m_y^2 + \frac{K_1}{4M^4} \left[ (m_x^2 - m_y^2)^2 + 4m_z^2 (m_x^2 + m_y^2) \right]$$

$$E^{(001)} = -H.M\frac{K_U}{M^2}m_z^2 + \frac{K_P}{M^2}m_y^2 + \frac{K_1}{4M^4} \left( m_x^2 m_y^2 + m_y^2 m_z^2 + m_z^2 m_x^2 \right) \tag{6.5-a,b}$$

where $K_U$ and $K_P$ are the uniaxial anisotropies directed in the film plane and normal to the film plane respectively and $K_1$ is the four-fold magnetocrystalline anisotropy.

The equations of motion are solved in the unprimed frame obtained from the transformation

$$\begin{pmatrix} m'_x \\ m'_y \\ m'_z \end{pmatrix} = \begin{pmatrix} \cos \phi & 0 & -\sin \phi \\ 0 & 1 & 0 \\ \sin \phi & 0 & \cos \phi \end{pmatrix} \begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix} \tag{6.6}$$

We derive expressions for the free energy in the unprimed co-ordinate system by substituting the primed variables in equation (6.5) with unprimed variables using equation 6.6. The effective
fields are calculated by retaining free energy terms to second order in small fluctuations in the magnetization around the equilibrium direction. Components of \( \frac{dm}{dt} \) in the unprimed frame are computed using equations 6.1, 6.4 and the transformation 6.6. For the (110) film we have

\[
\begin{align*}
\frac{dm_x}{dt} &= \gamma \left\{ H_0 \cos(\phi_H - \phi) + \left( \frac{2K_1}{M} \right) \left[ 1 - (2 \sin^2 \phi + \frac{3}{8} \sin^2 (2\phi)) \right] + \left( \frac{2K_U}{M} \right) \cos^2 \phi \\
&\quad + \left( \frac{2K_p}{M} \right) + \frac{2Ak^2}{M} \right\} m_y - \gamma Mh_y \\
\frac{dm_y}{dt} &= -\gamma \left\{ H_0 \cos(\phi_H - \phi) + \left( \frac{2K_1}{M} \right) \left[ 1 - \frac{1}{2} \sin^2 (2\phi) \right] + \left( \frac{2K_U}{M} \right) \cos^2 \phi + \left( \frac{2K_p}{M} \right) \cos^2 \phi \\
&\quad + \frac{2Ak^2}{M} \right\} m_x - \gamma Mh_x
\end{align*}
\]

while the components of \( \frac{dm}{dt} \) in the (001) oriented sample are given by

\[
\begin{align*}
\frac{dm_x}{dt} &= \gamma \left\{ H_0 \cos(\phi_H - \phi) + \left( \frac{2K_1}{M} \right) \left[ 1 - \frac{1}{2} \sin^2 (2\phi) \right] + \left( \frac{2K_U}{M} \right) \cos^2 \phi + \left( \frac{2K_p}{M} \right) \cos^2 \phi \\
&\quad + \frac{2Ak^2}{M} \right\} m_y - \gamma Mh_y \\
\frac{dm_y}{dt} &= -\gamma \left\{ H_0 \cos(\phi_H - \phi) + \left( \frac{2K_1}{M} \right) \left[ 1 - 2 \sin^2 \phi \right] + \left( \frac{2K_U}{M} \right) \left( 1 - 2 \sin^2 \phi \right) + \frac{2Ak^2}{M} \right\} m_x \\
&\quad + \gamma Mh_x
\end{align*}
\]

In the above equations \( A \) is the exchange stiffness and \( h_i \)'s \((i = x,y)\) are components of the dipolar fields. The exchange field is proportional to \( k^2 = k_x^2 + k_y^2 \), where \( k \) is the magnitude of the in-plane magnon wave-vector \((2k \sin \theta_i)\) and \( k_y \) is the wave-vector component perpendicular to the film. The sign conventions for the uniaxial anisotropies is chosen such that \( K_U > 0 \) puts an in-plane easy axis along [001] for the (110) film and along [100] for the (001) film. \( K_p \) is
defined such that $2K_p/M_0 + 4\pi M_0 < 0$ places an easy axis normal to the film plane, along $y$.

After linearization, the coupled equations for $m$ are solved together with the electromagnetic and exchange boundary conditions [15,21]. For a given frequency $\omega$ and in-plane wave-vector $q$, there are six solutions to $k_y$ for the above set of equations. Allowed solutions are superpositions of six partial waves corresponding to the $k_y$'s satisfying the boundary conditions appropriate to the film geometry.

The most striking effect of the dipolar interaction is the appearance of a surface localized excitation, $S$, the Damon-Eshbach mode. $S$ is most easily observed in light scattering experiments because of large dipolar fields near the film surface. Its frequency, $\omega_s$, also exhibits the interesting property of being largest for propagation perpendicular to $M_0$ and decreases as $q$ approaches the magnetization direction. In the latter case $S$ is no longer localized to the surface having transformed to the $n = 0$, uniform bulk mode. The (110) grown film provides direct evidence in zero applied field for such a transformation of the surface magnon thus confirming the direction of the easy axis of magnetization.

Figure 6.2 shows typical Brillouin spectra from the (110) oriented bcc Co film in the presence of a 2.0 kOe field for excitation at 5145Å. The spectra were recorded at an angle of incidence $\theta = 20^\circ$ with $\phi_1$ varying systematically between $0^\circ$ and $180^\circ$. The surface ($S$) and $n = 1$ bulk ($B_1$) magnons are clearly observed in the spectra. Figure 6.3 summarizes the field dependence of $\omega_s$ and $\omega_{h11}$. The difference between the surface and bulk frequencies is especially sensitive to the exchange constant ($A$) while the slope of the curves is controlled by
the gyromagnetic ratio \((\gamma)\). The solid curves through the data in figure 6.3 (and 6.4) are fits using the theory discussed above.

The two data sets for the surface mode in figure 6.3 (hollow and solid symbols) were taken using different magnet systems and Fabry-Perot interferometer calibrations. The slight differences are due to systematic variations in the two experimental setups. The solid line is the best fit made to both data sets. Data for the surface mode in figure 6.3 (hollow circles) extend all the way down to \(H_0 = 0\). The softening of the surface magnon frequency around \(H_0 = 1.2 \pm 0.1\) kOe is due to a competition between the external magnetic field and an in-plane uniaxial anisotropy, \(K_U\), that normally orients the magnetization along [001]. When the external magnetic field exceeds a critical field \(H_c\), the magnetization is pulled into the direction of the field. Simple free energy arguments can be used to estimate the critical field \(H_c\). If \(\alpha\) is the angle between \(H_0\) and \(M_0\), the total free energy in terms of the Zeeman and uniaxial anisotropy fields is given by (neglecting magnetocrystalline terms)

\[
E = -M_0 H_0 \cos \alpha + K_U \cos^2 \alpha
\]  

At equilibrium, the torques due to the two fields balance each other. The free energy is minimized by the condition \(\frac{\partial E}{\partial \alpha} = 0\). Hence,

\[
\sin \alpha (M_0 H_0 - 2K_U \cos \alpha) = 0
\]  

The critical field for a jump in the magnetization occurs when the free energy changes from a minimum to a maximum. This condition is given by \(\frac{\partial E}{\partial \alpha} = \frac{\partial^2 E}{\partial^2 \alpha} = 0\) [23]. The frequency of
the surface mode has a minimum at $H_c$ and thus provides a measure of $K_U$. Using the above relations it can be shown that $K_U = (H_c M_0)/2$.

The results of figure 6.4 indicate the absence of a four-fold magnetocrystalline anisotropy and instead reveal the presence of a large in-plane uniaxial anisotropy contribution to the surface and bulk magnon dispersion. The inset in figure 6.4 shows the behavior of $\omega_5$ measured at $\lambda = 6471 \text{Å}$ for directions $0^\circ \leq \phi_1 \leq 360^\circ$ using a 1.2 kOe magnetic field which is just enough to saturate the magnetization. On simultaneously fitting the data in figures 6.3 and 6.4 using the theory described above, the following results are obtained for the (110) grown film: gyromagnetic ratio $\gamma = 2.02$, exchange constant $A = 1.87 \times 10^6 \text{erg/cm}$, $K_P = 8.3 \pm 3 \times 10^5 \text{erg/cm}^3$ and $K_I = 0$ for the (110) film. Independently determined values of $4\pi M_0$ (13 kGauss) from d.c. SQUID measurements were used to fit the Brillouin data. The critical field $H_c (= 1.2 \pm 0.1 \text{ kOe})$ gives $K_U = 6.2 \pm 0.5 \times 10^5 \text{erg/cm}^3$ (Table 6.1). The magnitude of $K_U/M$ largely determines the difference in the magnon frequencies between the easy and hard directions that occur at $\phi_1 = 0^\circ$ and $\phi_1 = 90^\circ$. With all other parameters held constant, the quantity $4\pi M_0 + 2K_P/M_0$ influences the mean value of $\omega_5$ and $\omega_{11}$ between $0 < \phi_1 < 180^\circ$. The larger error in $K_P$ reflects the insensitivity of $\omega_5$ and $\omega_{11}$ to $K_P$ because $4\pi M_0$ is more than a factor of ten larger than $2K_P/M_0$. The same values of each of the anisotropies ($K_U$ and $K_P$) are used to fit both the bulk and surface magnon in-plane dispersions (i.e. $\omega_5$, $\omega_{11}$ vs $\phi_1$) which is consistent with the idea that these uniaxial anisotropies are indeed bulk values. This is significant because $\omega_{11}$ could be more sensitive to surface anisotropies than $\omega_5$ through pinning effects. Hence, we
conclude that there is little evidence for surface anisotropies being different from bulk anisotropies in the (110) Co film.

Figure 6.5 shows illustrative spectra of the surface magnon from the 216Å (001) grown film for $\mathbf{q}$ along various crystal directions. The top portion of the figure shows a spectrum taken at a larger free spectral range that extends out to the first exchange dominated bulk mode also. As in the (110) film, the bulk mode intensity is extremely weak compared to the surface mode. The slight asymmetry observed in the Stokes/anti-Stokes bulk mode intensity which is possibly similar to the case of Fe films discussed in chapter V. This feature was not analyzed in detail as the bulk mode signal are very weak. The in-plane anisotropy is barely evident for different propagation directions in this film. Figure 6.6 summarizes the $\omega_S$ vs $H$ and $\omega_S$ vs $\phi_1$ data from the (001) grown Co epilayer. The magnon frequencies for this (001) grown sample surprisingly exhibits a two-fold symmetry and the data reveals that the two in-plane [100] and [010] directions are magnetically inequivalent. However, since the $\omega_S$ along these two crystallographically equivalent directions differ by only about 1 GHz, the uniaxial anisotropy is much smaller than in the (110) film. The bulk mode is not included in the analysis because the error in the measurement of the bulk mode frequency is of the order of the in-plane anisotropy.

The anisotropy values deduced from fits to the Brillouin data in figure 6.6 show that $K_U$ for this film is indeed an order of magnitude smaller ($0.8\pm0.2x10^3$ erg/cm$^3$) than that in the (110) grown sample (table 6.1). $K_U$ cannot be accurately determined from the surface mode
softening (as in $\omega_\delta$ vs $H$ data of the (110) film, figure 6.3) because the uniaxial anisotropy is much smaller in this case. The value of $4\pi M_0$ used for the Brillouin calculation (13.5 kGauss) was determined by independent d.c. SQUID measurements. Other parameters obtained from the fits were $A = 1.9 \times 10^{-6} \text{ erg/cm}^2$, $\gamma = 2.02$ and $K_F = -3.7 \pm 2 \times 10^5 \text{ erg/cm}^3$. A small $K_1 (-0.2 \times 10^3 \text{ erg/cm}^3)$ utilized in this fit reproduces the flattening of the in-plane dispersion evident at $0^\circ \leq \phi_1 \leq 30^\circ$ and at $150^\circ \leq \phi_1 \leq 180^\circ$ in figure 6.6(2).

The value of $K_U$ in the (110) film obtained from the spin wave softening was also confirmed using d.c. SQUID measurements. Figure 6.7 shows the $M$ vs $H$ data for the (110) film. For $H$ along the easy axis, $M$ switches direction sharply with the reversal of $H$. When $H$ is along the hard axis, a larger field is required to saturate the magnetization due to the presence of $K_U$. The saturation field $H_s (1.25 \pm 0.1 \text{ kOe})$ and magnetization $4\pi M_0 = 13 \text{ kGauss}$ provides a direct measure of $K_U (= 6.5 \pm 0.5 \times 10^5 \text{ erg/cm}^3)$ which agrees with the result from Brillouin spectroscopy. The critical field of the (001) film could not be determined from SQUID measurements as the anisotropy in the film is very small.

Independent evidence for the orientation of the spontaneous axis of magnetization in each of the films was obtained by studying the directional dependence of the surface magnon. Figure 6.8 shows this behavior in the (110) Co film in zero applied field. The in-plane direction of the magnon wave-vector $q$ is denoted by the angle $\phi_q$ measured from the $[\bar{1}10]$ crystal direction. When $q$ is directed along $[\bar{1}10]$ ($\phi_q = 0^\circ$), a large asymmetry is evident in the Stokes/anti-Stokes (S/aS) intensity ratios. However, as $q$ approaches [001] ($\phi_q = 90^\circ$) the S/AS
intensity asymmetry weakens indicating a loss of surface-like character. The S/aS intensity ratio shows a sudden drop around $\phi_\perp = 75^\circ$ where the surface mode becomes delocalized and appears bulk-like. When $q \parallel [001]$ (easy axis, $\phi_\perp = 90^\circ$), the surface magnon transforms into the $n = 0$ bulk mode ($B_0$). As $q$ moves away from $[001]$ ($\phi_\perp = 105^\circ$ and $120^\circ$) the S/aS intensity ratio switches with the Stokes peak now being the weaker of the two. The intensity of the surface mode at $\phi_\perp = 60^\circ$ on the Stokes side is approximately the same as the intensity at $\phi_\perp = 120^\circ$ on the anti-Stokes side, thus indicating that the magnetization is oriented midway at $\phi_\perp = 90^\circ$ along [001].

Figure 6.9 shows calculated Brillouin intensities for this mode on the (110) film as a function of $\phi_\perp$. The scattering intensity (thick film geometry) has the form [17,18,24,25]

$$ I = r_{yy} S_{yy}(\omega, q) + r_{xx} S_{xx}(\omega, q) + 2 \text{Re}( r_{xy} S_{xy}(\omega, q) ) $$

where the $r$'s depend on the index of refraction, polarization and the angle of incidence of the probing electromagnetic wave. The amplitude of the thermal spin fluctuations are given by components of the spin correlation function $S(\omega, q)$. The $r$'s are in general complex in the case of an absorbing medium described by a complex index of refraction. Because of this, the $r_{xy}$ term is non-zero and responsible for a large S/aS asymmetry through the $S_{xy}$ component of the correlation function. The spin correlation functions are constructed from the solutions $\psi(\omega, q, \gamma)$ that satisfy the equations of motion and the associated boundary conditions at frequency $\omega_\perp$ [24,25]. The intensities plotted in figure 6.9 were calculated using the parameters listed in table
6.1. For $H = 0$ (as in figure 6.8), the calculated $S/aS$ intensity ratios show the same general behavior found in the experiment.

4 Discussion

The Brillouin data from both the bcc Co films indicated the presence of uniaxial anisotropies. The origin of uniaxial anisotropies observed in these cubic films is determined in this section. We consider various possible contributions to the total effective anisotropy used to fit the spin wave dispersion in order to determine the origin of uniaxial anisotropies in our spin wave data. The effective magnetic anisotropy energy ($K_{\text{eff}}$) is a combination of the demagnetization ($K_D$), magnetocrystalline ($K_{\text{MC}}$), magnetoelastic ($K_{\text{ME}}$) and surface anisotropy ($K_S$) energies; i.e. $K_{\text{eff}} = K_D + K_{\text{MC}} + K_{\text{ME}} + K_S$. In our convention, $K_{\text{eff}}$ is positive for an in-plane easy axis. The demagnetization energy for a thin film is $K_D = 2\pi M_0^2$, where $M_0$ is the magnetization per unit volume of bcc cobalt. Fits to the Brillouin data reveal $K_1 = 0$ for the (110) film and hence there is no contribution to $K_{\text{eff}}$ from the fourth order magnetocrystalline anisotropy. Due to the relatively large thickness of the samples used in this study, surface anisotropies ($K_S$) should not be significant. Moreover, the Brillouin fits provide no evidence for surface anisotropies in the (110) film and hence we set $K_S = 0$. Thus, in the absence of a magnetoelastic contribution we obtain $K_{\text{eff}}[110] = K_{\text{eff}}[001]$. This is inconsistent with the observation of a preferential orientation of the in-plane magnetization along [001] for the (110) grown film.
A weak $K_1$ is present in the (001) film. However the cubic symmetry of this term forbids a uniaxial contribution to $K_{\text{af}}$ along either principal axis in this film. Hence in this case also, contributions from $K_S$ and $K_{\text{MC}}$ to the free energy cannot explain the 1 GHz difference between the measured spin wave frequencies along the two principal crystal axes. In what follows, we consider the possibility that the uniaxial anisotropies for both orientations are primarily due to magnetoelastic contributions.

The magnetoelastic contributions to $K_{\text{af}}$ are calculated along the lines followed in [6,7,8] (appendix C). The geometry is chosen so that the $y$ axis is normal to the film plane and the $zx$ axes lie in-plane along the easy and hard axes of magnetization respectively. The magnetoelastic energy for the (110) oriented film is given by $K_{ME} = -\sum_y \alpha_i^2 b_y e_j$ where $b_y$'s are elements of the body transformed magnetoelastic tensor, $e = (e_x, e_y, e_z)$ are the diagonal components of the strain and $\alpha^2 = (\alpha_x^2, \alpha_y^2, \alpha_z^2)$ are squares of the direction cosines. $e_y$ is the out-of-plane strain and $(e_x, e_z)$ lie in-plane.

\[ K_{ME} = -\alpha_x^2 [(b_{11} - b_{12})(e_x - \frac{e_x + e_y}{2}) + b_{44}(e_y - e_x)] - \alpha_y^2 2b_{44}(e_y - e_x) \]

(6.12)

In deriving the above expression the $\alpha_x^2$ term has been eliminated through the relation $\sum_{i=1,3} \alpha_i^2 = 1$. The strains are evaluated by minimizing the elastic free energy

\[ F(e) = \frac{1}{2} \sum_{ij} e_i C_{ij} e_j \]

with respect to the out-of-plane strain $e_y$, where $C_{ij}$'s are the elasticity tensor elements in the transformed co-ordinate frame. Minimizing $F(e)$ yields
\[ e_y = \left( -\frac{1}{a_{11}} \right) \left[ a_{12} e_x + c_{12} e_z \right] \]  

(6.13)

where \( a_{11} = \left[ (c_{11} + c_{12})/2 + c_{44} \right] \) and \( a_{12} = \left[ (c_{11} + c_{12})/2 - c_{44} \right] \). Since bcc Co is closely lattice matched with the substrate, the in-plane strains arising from the predicted film-substrate lattice mismatch are very small; \( e_x \approx e_z = 0.25\% \) [3]. Together with previously measured \( C_{ij} \)'s of the Co films [14], we obtain \( e_y = -0.33\% \).

In order to estimate the magnetoelastic energy, the magnetoelasticity tensor elements \( b_{ij} \) are required. Since this data is not available for bcc cobalt, values for fcc Co derived from measured magnetostriction constants, \( \lambda_{100}, \lambda_{111} \) of Co-rich fcc Co/Pt alloys are used instead [26]. Beyond a concentration of 50% Co, the \( \lambda \)'s for the alloy do not change appreciably and hence are reasonable values for the magnetostriction constants of fcc Co.

\[
\lambda_{100} = \left( -\frac{2}{3} \right) \left[ \frac{b_{11} - b_{12}}{c_{11} - c_{12}} \right], \quad \lambda_{111} = -\left[ \frac{b_{44}}{3c_{44}} \right] 
\]

(6.14)

Using values of \( \lambda_{100} (= 130 \times 10^{-6}) \), \( \lambda_{111} (= -65 \times 10^{-6}) \) we compute the combinations of \( b_{ij} \)'s essential to evaluate the magnetoelastic free energy and obtain

\[
K_{\text{MIE}} = \alpha_x^2 (6.7 \times 10^5) + \alpha_y^2 (10.8 \times 10^5) \text{ erg/cm}^3
\]

(6.15)

The magnetoelastic energy thus gives rise to both an in-plane and a perpendicular anisotropy in the (110) oriented case. The computed in-plane (coefficient of \( \alpha_x^2 \)) and out-of-plane (coefficient of \( \alpha_y^2 \)) anisotropies agree well with the \( K_U \) and \( K_P \) values deduced from fits to the Brillouin data (\( K_U = 6.2 \pm 0.5 \times 10^5 \text{ erg/cm}^3 \) and \( K_P = 8.3 \pm 3 \times 10^5 \text{ erg/cm}^3 \), table 6.1).

A similar calculation of the magnetoelastic energy for the (001) oriented film yields
\[ K_{\text{ME}} = [\alpha_z^2 (e_x - e_z) + \alpha_y^2 (e_x - e_y)](b_{11} - b_{12}) \] 

(6.16)

As before, \( e_y \) is the out-of-plane strain while \((e_z, e_x)\) are the in-plane strains along the easy and hard axes respectively. Minimizing the elastic free energy with respect to \( e_y \), omitting shear strains gives \( e_y = -\left(\frac{c_{12}}{c_{11}}\right)(e_x + e_z) \). After substituting values of strains and magnetoelastic constants, we obtain \( K_{\text{ME}} = \alpha_z^2(0) + \alpha_y^2(-3.1 \times 10^5) \) erg/cm\(^3\). The corresponding results from Brillouin scattering are \( K_U = (0.8 \pm 0.2) \times 10^3 \) erg/cm\(^3\) and \( K_P = (-3.7 \pm 2) \times 10^3 \) erg/cm\(^3\). Once again good agreement is obtained between the BLS results and magnetoelasticity calculations.

If the two in-plane strains \( e_x \) and \( e_z \) are strictly equal there is no contribution from magnetoelastic factors to \( K_U \) for the (001) oriented film. On the other hand slight differences between the in-plane strains would give rise to a finite magnetoelastic contribution, as observed in the small difference (1 GHz) between the magnon frequencies measured along the two crystallographic directions. The magnitude of \( K_U \) for the (001) film is almost an order of magnitude smaller than that for the (110) sample. This is consistent with the small (1 GHz) difference in \( \omega_b \) observed in the (001) film as opposed to a larger anisotropy (6 GHz) seen in the (110) film, for \( \mathbf{q} \) along the two principal in-plane directions in the respective samples. Finally, we note that the small value of \( K_U \) in the (001) film is equivalent to an effective interfacial anisotropy of 0.16 erg/cm\(^2\). While this value is a reasonable estimate for a surface anisotropy in ferromagnetic thin films [13], we are unable to determine whether \( K_U \) for the (001) grown sample is a surface or bulk anisotropy. This is because the bulk magnon in-plane
dispersion was not included in the analysis as the error in measured bulk magnon frequency was comparable to the in-plane anisotropy (~1 GHz).

The above calculations were performed assuming an isotropic in-plane strain. This condition is valid when the primitive lattice vectors of the film and substrate vary only by a scale factor and the angle between them is identical [27-29]. BCC Co on GaAs satisfies this condition. Recently bcc Co was also stabilized on a tetragonal TiAl substrate and the in-plane isotropic strain condition is not satisfied in this case [30]. The lattice constants along the principal crystal directions are required to compute all the interface strains.

Before concluding this section we call attention to the values of magnetization used in this study. The average measured value of $4\pi M_0$ for the two films is about 13.2 kGauss corresponding to an average magnetic moment ($\mu$) of 1.3$\mu_B$/atom. In contrast, the measured $\mu$ in a thicker (357Å) bcc Co film [3] was over 1.5$\mu_B$/atom while the theoretically predicted magnetic moment is 1.7$\mu_B$/atom [31]. The reason for the reduction in $\mu$ for thinner films is not clear. A recent calculation proposed that arsenic diffusion from the substrate suppresses the average magnetic moment of bcc Co [32]. Evidence for a lower local magnetic moment per atom in bcc Co films has also been reported from neutron scattering measurements [33]. This study indicated that $\mu$ varied significantly with depth within a 100Å film. While the magnetic moment at the center agreed with the theoretically predicted value of 1.7$\mu_B$ per atom, it reduced to 1.0$\mu_B$ per atom at the surfaces which yields an average of about 1.3 -1.4$\mu_B$ per atom for the entire film. If we assume that a similar reduction in magnetic moment occurs in
our 200Å thick cobalt samples it would explain the reduced values of $M_0$ obtained from SQUID measurements.

5 Summary

The in-plane spin wave dispersion was measured in bcc cobalt epilayers of different orientations using Brillouin light scattering. In the (110) oriented film we find a large in-plane two-fold uniaxial anisotropy while in the (001) film, a similar, though much weaker anisotropy is observed. Free energy considerations show that magnetoelastic interactions, due to lattice mismatch strains at the film-substrate interface can give rise to the uniaxial anisotropy observed in the (110) oriented epilayer. The value of the anisotropy constants deduced from these energy considerations using reasonable estimates for magnetoelastic constants and strains are found to agree with those deduced from fits to the spin wave frequencies. The easy axis of magnetization is also directly evident by the transformation of the surface magnon into the $n = 0$ bulk magnon when the magnon wave-vector lies parallel to the spontaneous (easy) axis of magnetization. The much weaker uniaxial anisotropy measured for the (001) oriented Co-layer is also consistent with magnetoelastic effects although, for exact tetragonal in-plane strain, a two-fold anisotropy should be completely suppressed. The observed uniaxial anisotropy in the (001) oriented film hence suggest that if indeed the in-plane strains are equal (i.e. $e_x = e_z$) then an additional, presently unknown, mechanism could also contribute to the small differences in the spin wave frequencies along [100] and [010]. The need for a mechanism not related to
magnetoelastic effects was suggested by recent observations of uniaxial anisotropies in Fe_{1-x}Co_{x}
alloys, evident even at alloy concentrations that provided an exact lattice match between
epilayer and substrate [34]. Thus while magnetoelastic effects likely provide the most
significant contribution to magnetic anisotropies in the stabilized phase of bcc cobalt, other
causes may be required to account for the residual anisotropies evident in the (001) Co film.
Figure 6.1: Co-ordinate system for spin wave dispersion calculation. The $y'$ axis is normal to the film and the $(x', z')$ axes lie in the film plane. The film is bounded by the planes $y' = 0, h$. The unprimed reference frame is obtained by a rotation of $\phi$ about $y' = y$. The magnetization is aligned along $z$. The angle between $M$ and $H$ is $(\phi_{\text{H}} - \phi)$. 
Figure 6.2: Depolarized p → s Brillouin spectra from (110) grown 202 Å thick bcc Co film in backscattering, at an angle of incidence $\theta_i = 20^\circ$, $H = 2.0$ kOe, $\lambda = 5145$ Å. In all experiments, $q \perp H$ ($\phi_H = 0^\circ$) corresponds to $q$ propagating along [\bar{1} 10] while $\phi_H = 90^\circ$ refers to $q$ along [001]. S identifies the surface magnon and $B_1$ the first order bulk magnon.
Figure 6.3: Magnetic field dependence of the surface ($\omega_S$) and bulk ($\omega_{B1}$) magnon frequencies in the (110) oriented film. The solid curve through the data is a theoretical fit as discussed in text. The error bars on $\omega_S$ and $\omega_{B1}$ are ±0.5 GHz and ±1.0 GHz respectively. The error in the measured magnetic field $H$ is ±0.1 kOe.
Figure 6.4: In-plane magnon dispersion at two different excitation wavelengths from (110) oriented Co-film. The main figure shows spin wave frequencies measured at 5471Å while the inset shows data recorded with 6471Å radiation. The solid lines through the surface ($\omega_S$) and bulk ($\omega_{B1}$) magnon frequencies are a fit utilizing parameters discussed in the text. The error bars on $\omega_S$ and $\omega_{B1}$ are ±0.5 GHz and ±1.0 GHz respectively. The error in $\omega_S$ at 6471Å is ±0.2 GHz. The angle $\phi_1$ denotes the in-plane angle between the crystallographic [001] direction and the external magnetic field and the error in $\phi_1$ ($\Delta\phi_1$) is ±3°.
Figure 6.5: Brillouin spectra from surface magnon in (001) grown 216Å film. 1. Bulk mode and surface mode 2. Surface modes for a series of angle $\phi_1$. The in-plane anisotropy in this film is much smaller than the (110) grown film.
Figure 6.6: Field dependence and in-plane dispersion of surface magnon shown for the 216Å thick, (001) oriented Co epilayer. 1. $\theta_i = 30^\circ$, $H \parallel [100]$, $q \perp H$, $\lambda = 5145\text{Å}$. The error bars on $\omega_s$ is ±0.5 GHz. 2. $\theta_i = 60^\circ$, $H = 1.2 \text{kOe}$, $q \perp H$, $\lambda = 5145\text{Å}$. Since the in-plane anisotropy is very small the measurements were performed at a higher resolution (< 0.1 GHz).
Figure 6.7: D.C. SQUID data from (110) grown film. When $H$ is along the hard ([1 1 0]) axis, a large saturation field is required. This field provides a direct measurement of in-plane uniaxial anisotropy. The magnetization $M_0$ is obtained by dividing the total magnetic moment ($22 \times 10^5$ emu/cm$^3$) by the sample volume (for 202 Å film, area = 0.105 cm$^2$, volume $V = 2.12 \times 10^{-7}$ cm$^3$). The measured saturation magnetization $M_0 = 1035$ Gauss gives $4\pi M_0 = 13$ kGauss.
Figure 6.8: In-plane dispersion, measured for $\theta = 60^\circ$, $\lambda = 5145\text{Å}$, $H = 0$ for (110) film. The backscattered light was not analyzed to retain both the Rayleigh phonon (R) and the surface magnon (S) features. At $\phi = 75^\circ$ and $\phi = 82^\circ$ (direction of $\mathbf{q}$ from [1 1 0]), the intensity of the anti-Stokes spectrum (identified by *) shows a significant increase. When $\phi = 90^\circ$, i.e. for $\mathbf{q}$ along [0 0 1] and parallel to $\mathbf{M}_0$, the surface mode peaks are replaced by two equal intensity, $n = 0$, bulk magnons ($\mathbf{B}_0$) peaks.
Figure 6.9: Calculated Brillouin spectra for the (110) oriented 202Å Co film as a function of the in-plane angle $\phi_q$. As $q$ approaches [001] (i.e. $\phi_q \to 90^\circ$) the Stokes/anti-Stokes ratio becomes equal while at angles away from this direction there is a large asymmetry in the magnon intensities indicative of surface-like behavior.
Table 6.1: Anisotropy parameters of bcc Co.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Anisotropy x 10^3 erg/cm^3</th>
<th>Brillouin Experiment</th>
<th>Magnetoelastic Calculation</th>
</tr>
</thead>
<tbody>
<tr>
<td>(110) film</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kn</td>
<td>6.2 ± 0.5</td>
<td>6.7</td>
<td></td>
</tr>
<tr>
<td>Kp</td>
<td>8.3 ± 3.0</td>
<td>10.8</td>
<td></td>
</tr>
<tr>
<td>K1</td>
<td>0.0</td>
<td>NA</td>
<td></td>
</tr>
<tr>
<td>(001) film</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kn</td>
<td>0.8 ± 0.2</td>
<td>0.0</td>
<td></td>
</tr>
<tr>
<td>Kp</td>
<td>-3.7 ± 2.0</td>
<td>-3.1</td>
<td></td>
</tr>
<tr>
<td>K1</td>
<td>-0.2</td>
<td>NA</td>
<td></td>
</tr>
</tbody>
</table>

Summary of uniaxial anisotropies obtained from Brillouin light scattering experiments and magnetoelastic energy calculations. $K_n$ and $K_p$ are the in-plane and perpendicular uniaxial anisotropies respectively. Other parameters extracted from the fitting procedure for the (110) film are $A = 1.87 \times 10^{-6}$ erg/cm, $\gamma = 2.02$. For the (001) film, $A = 1.9 \times 10^{-6}$ erg/cm, $\gamma = 2.02$.

SQUID measured values of $4\pi M_0$ are 13.0 and 13.5 kGauss in the (110) and (001) films respectively. The magnetization $M_0$ is obtained by dividing the total magnetic moment by the sample volume (for 202Å film, magnetic moment = $22 \times 10^{-5}$ emu/cm$^3$, area = 0.105 cm$^2$, volume $V = 3.01 \times 10^{-7}$ cm$^3$; for 216Å film, magnetic moment = $32 \times 10^{-5}$ emu/cm$^3$, area = 0.139 cm$^2$, volume $V = 2.12 \times 10^{-7}$ cm$^3$).
References


CHAPTER VII

Optical Properties of GaAs/AlGaAs Quantum Wells

This chapter discusses the electronic and optical properties of GaAs/AlGaAs quantum well heterostructures. The concept of excitons and their role in determining the optical properties of quantum well devices is addressed.

1 Electronic Properties

The direct bandgap and high electron mobilities of materials like GaAs make them excellent candidates for high frequency opto-electronic and microwave device applications [1-4]. These systems also offer an excellent platform to study the quantum effects of particle confinement [5-10]. The explosive progress in this field is due to the emergence of molecular beam epitaxy as a growth technique where quantum well growth can be controlled at the atomic monolayer level [11,12].

III-V binary compounds like GaAs have 8 outer orbit electrons that contribute to the electronic properties of the material. The outer shell orbital electrons (s and p like) of all atoms hybridize to form bonding and anti-bonding orbitals. The orbital levels broaden into bands due to the large number of unit cells in the crystal. The bonding s-like orbitals are deep levels and are always occupied by 2 electrons per unit cell. The remaining 6 electrons fill the three bonding p orbitals to form the valence band. The bands originating from anti-bonding orbitals are empty and the lowest lying one (usually s-like) forms the conduction band [10]. The top of the valence band occurs at center of the Brillouin zone
In the absence of spin-orbit coupling this band is 6-fold degenerate and constitutes the valence band. Spin-orbit coupling lifts the 6-fold degeneracy and gives rise to a quadruplet corresponding to $J = 3/2$ and a doublet corresponding to $J = 1/2$. The 4-fold degenerate valence are called the heavy hole and light hole bands according to the band curvatures that determine the effective mass of the carriers. Since the anions originate from the $p$ orbitals of the anion, the spin-orbit coupling is primarily a property associated with the anion.

The conduction band minimum is often found at the $\Gamma$ point in III-V materials but it can also occur at the $X$ ([001]) or $L$ ([111]) point. In general, the heavier the cation, the more likely it is to have a $\Gamma$ point minimum. Further, the valence band - conduction band separation at the $\Gamma$ point (known as the bandgap, $E_g$) varies inversely as the cation mass. All these trends can be understood in terms of the ionicity of the bond between the two chemical species of the compound semiconductor [14].

Figure 7.1 shows the band diagram of GaAs around the zone center. GaAs is a direct bandgap III-V semiconductor with a zinc blende crystal structure. Bulk GaAs has a light hole and a heavy hole valence band that are degenerate at $k = 0$ ($\Gamma_v$ band), and a spin-orbit split off band ($\Delta$, $\Gamma_7$ band) that lies 0.34 eV below the valence band. The conduction band minimum lies 1.42 eV above the valence band maximum at $k = 0$ (energy bandgap) at 300 K. Other conduction band minima occur at the zone boundaries along the $[110]$ ($L$ valley) and the $[111]$ ($X$ valley) directions. The $L$ and $X$ valley minima are at 0.29 eV and 0.48 eV respectively above the $\Gamma$ valley minimum.

AlAs is an indirect bandgap III-V semiconductor with a $\Gamma$ point bandgap of 3.02 eV and a spin-orbit split off band 0.3 eV below the valence band. However, the alloy $\text{Al}_x\text{Ga}_{1-x}\text{As}$ has a direct bandgap minimum at the $\Gamma$ point for $x \leq 0.4$. Electronically,
Al$_x$Ga$_{1-x}$As is not a crystalline structure because there is no translational invariance in the potential experienced by the electrons, due to the random distribution of Al and Ga atoms at various lattice sites. However, an average lattice potential can be defined in a mean field approximation and represented as a weighted average $<V> = x<V_{Al}> + (1-x)<V_{Ga}>$. This procedure restores translational invariance and Bloch waves, effective masses, etc. may be defined for Al$_x$Ga$_{1-x}$As as in case of a binary alloy. An advantage of using ternary alloys is that many material properties (like bandgap) can be varied smoothly by changing the Al concentration $(x)$ [6,10]. A detailed band structure calculation of GaAs near the center of the Brillouin zone has been performed using the Kane model [10,15-17].

2 Optical Properties

The absorption coefficient of a medium is calculated from the interaction of the electromagnetic field with the charge carriers in the medium. The single electron interaction Hamiltonian is given by [10]

$$H = H_0 + \frac{e}{2mc}(p \cdot \vec{A} + \vec{A} \cdot p)$$

(7.1)

The electromagnetic field is described by a monochromatic plane wave of frequency $\omega$

$$E = E_0 \phi \cos(\omega t - k \cdot r) \quad \text{and} \quad \vec{E} = -\frac{1}{c} \frac{\partial \vec{A}}{\partial t}$$

(7.2)

where $\phi$ is the polarization of the electromagnetic field. The Fermi golden rule gives the transition probability from the initial state $|i\rangle$ to the final state $|f\rangle$

$$\tilde{P}_{i,o} = \frac{2\pi}{\hbar} |<f|V|i>|^2 \delta(E_f - E_i - \hbar \omega)$$

(7.3)

where $V = \frac{ieE}{4m_0\omega} \cdot \vec{e} \cdot \vec{p}$ in the dipole approximation. The final transition probability depends on the Fermi occupancy factors of the valence and conduction bands.
\[ P_{i \rightarrow f} = \bar{P}_{i \rightarrow f} f(E_i)(1 - f(E_f)) \]  

(7.4)

The difference between the upward and downward transition rate yields the net absorption coefficient. The net energy loss of the electromagnetic wave per unit time associated with the \(|i\rangle \rightarrow |f\rangle\) transition is \(\hbar \omega [P_{fi} - P_{if}]\) and the resulting absorption coefficient is

\[
\alpha(\omega) = A \sum_{i,f} |\tilde{M}_{if}|^2 \delta[E_f - E_i - \hbar \omega] [f(E_i) - f(E_f)]
\]  

(7.5)

where \(A\) is a constant and \(M_{if}\) is the dipole matrix transition element given by \(M_{if} = \langle i|p|f\rangle\). Figure 7.2 shows the absorption spectrum of bulk GaAs. The fundamental absorption edge in GaAs arises from direct band to band transitions. Photons with energies greater than the bandgap of GaAs are absorbed by valence band electrons that get excited into the conduction band. In the parabolic band approximation, energy conservation gives

\[ \hbar \omega = E_f = E_g + \frac{\hbar^2 k^2}{2} \left( \frac{1}{m_e} + \frac{1}{m_h} \right) \]  

(7.6)

where \(\hbar \omega\) is the photon energy, \(m_e, m_h\) are the electron and hole masses respectively. The hole mass refers to either the heavy or the light hole mass depending on the transition. Conduction band electrons suffer collisions with ions or other charge carrier and fall back into the valence band. Radiation emitted in this process is known as photoluminescence (PL) and it usually occurs at a slightly lower energy than absorption because some energy is lost in the collision process. Both absorption and photoluminescence are direct transitions between conduction and valence band states where the net momentum change in the process is close to zero; i.e. \(\Delta k \approx 0\). Indirect transitions where \(\Delta k \neq 0\) involve the participation of a phonon that carries away the excess momentum.

The shape of the absorption step in 3-D has the form \(\sqrt{\hbar \omega - E_g}\) due to the \(\sqrt{E}\) dependence of the 3-D density of states. However, a marked peak appears below the
absorption edge which is due to the excitonic contributions. Excitons are many body electron-hole states that are correlated through Coulomb interactions [18-20]. An electron in the conduction band interacts with an hole in the valence band via Coulomb interactions and forms a *stable bound state* with energy

\[ E_x = E_v + E_e - \frac{e^2}{\varepsilon r} \]  

(7.7)

where \( E_e \) and \( E_v \) are the energies of the conduction band electron and the valence band hole respectively and \( \varepsilon \) is the dielectric constant of the medium. Hence, the exciton energy \( E_x \) is lower than the free electron-hole energy by the Coulomb interaction term \( <e^2/\varepsilon r> \), which results in a sharp absorption peak below the band edge. The \( \Delta k = 0 \) selection rule is valid in the case of excitonic absorption also. In PL studies, the many body nature of excitons can be ignored and they may be treated like hydrogen atoms with a reduced effective mass. Many body effects become important only at high carrier densities.

Depending upon the dimensionality of the problem, appropriate single particle eigenstates and eigenvalues are constructed. In bulk GaAs, the exciton binding energy using a simple hydrogen-like model is given by

\[ E^{3D}_a = \frac{m^* e^4}{2\hbar^2 \varepsilon^2 n^2} \]  

(7.8)

where \( m^* = \frac{1}{m_e} + \frac{1}{m_h} \)^{-1} is the reduced mass of the system. After substituting values for the GaAs parameters [6], we get binding energy \( E^{3D}_a \approx 4.2 \text{ meV} \) for the heavy hole exciton [9]. Excitonic effects are therefore manifest only at low temperatures in bulk GaAs. The theory of excitonic absorption in semiconductors is a complicated calculation due to the many body nature of the exciton state and was first developed by Elliott [18]. Applications specific to III-V semiconductors are described in [20].
3 Confinement Effects

Quantum wells are fabricated by sandwiching a lower bandgap material like GaAs between a larger bandgap material such as AlGaAs. Figure 7.3 shows an AlGaAs/GaAs/AlGaAs heterostructure accompanied with an energy band diagram. AlGaAs provides square well potential barriers that confine electrons and holes in the GaAs layer. The carriers are free to move laterally within the GaAs layer (well) but their motion is constrained in the growth direction (usually labeled z) by the AlGaAs barriers. The well thickness typically ranges from 5 to 100 monolayers of GaAs (1 monolayer = 2.83Å) while the barriers are about 200-300Å wide. Quantum well samples with relatively few defects yield a highly mobile electron/hole gas that is confined to nearly two dimensions. The electron/hole confinement potential can be controlled by varying the Al (x) concentration in the barrier. However, lattice match criteria for strain free epitaxial growth require that x \leq 0.4. Wave functions describing carriers in a quantum well are essentially the same as those for a particle in a box. In the infinite potential barrier approximation, the wave functions take the form

$$\Psi(r, z) = U(K_{||}, z) \exp(iK_{\perp}r) \sin(k_{z} z)$$

where \( r \) is the in-plane radius vector, \( K_{||} \) is the in-plane wave-vector, \( k_{z} \) is the wave-vector component normal to the film which becomes \( (n_{d}L_{z}) \) in the infinite potential barrier approximation (\( n \) is the principal quantum number of the well state). \( L_{z} \) is the width of the quantum well and \( U(K_{||}, z) \) is the cell periodic Bloch wave function. The form of the wave functions determines the selection rules for absorption and photoluminescence. In the infinite barrier case only transitions between electron and hole states that satisfy \( n_{e} - n_{h} = 2n \), where \( n_{e} \) (\( n_{h} \)) are the principal quantum number of the electron (hole) states. Though \( n \) could take any value, only the \( n = 0 \) transition is readily observed from PL experiments in
quantum wells as the intensities of the higher order transitions are extremely weak and fall as $1/(n + 1)^3$ [2,21]. The $n \neq 0$ transitions appear as weak high energy shoulders in PL experiments.

Every well can support at least one bound electron and hole state. The maximum number of bound states depends on the depth of the potential barrier and is given by [10]

$$N(L_z) = 1 + \text{Int} \left[ \frac{2m^* V_0 L_z^2}{\pi^2 \hbar^2} \right]$$

(7.10)

where $V_0$ is the depth of the well. An important effect of confinement is the removal of heavy hole-light hole valence band degeneracy at $k = 0$ [22,23]. In GaAs, the light and heavy holes have masses of $0.082m_0$ and $0.45m_0$ respectively while the conduction band electron has a mass of $0.067m_0$ where $m_0$ is the bare electron mass. The energy levels of the quantum well are like those of a particle in a box

$$E = E_s + \frac{\hbar^2 \pi^2 n^2}{2m^* L_z^2}$$

(7.11)

where $m^*$ is the reduced electron-hole mass. The exciton PL energies in quantum wells in the infinite potential barrier approximation is given by

$$E_x = E_s + \frac{\hbar^2 \pi^2 n^2}{2m^* L_z^2} - E_b^{2D}$$

(7.12)

where $E_b^{2D}$ is the 2-D exciton binding energy.

Figure 7.4 shows the absorption spectrum from a series of GaAs/Al$_{0.2}$Ga$_{0.8}$As multiple quantum well samples. The top graph shows a 4000Å thick (bulk-like) sample that shows a single exciton absorption peak at the band edge. The thinner samples show several excitonic peaks corresponding to transitions between various quantized levels in
the wells. The lower graph in figure 7.4 shows the well width dependence of the exciton energies. The dots are data points and the solid lines are calculated energies. Although the thicker wells exhibit a $1/L^2$ behavior, equation 7.6 cannot describe the energy levels in the thinner wells where the finite depth of the potential wells must be taken into account.

Excitons play a very important role in determining the optical properties of quantum wells. Electron and hole wave function overlap strongly due to confinement and this modifies the absorption and PL properties of the wells [1-3]. 2-D confinement also enhances the ground state exciton binding energy (by a factor of 4) [21]. When excitons are modeled like 2-D hydrogen atoms, the binding energy has the form

$$E_{n}^{2D} = \frac{m^* e^4}{2\hbar^2 e^2 (n + \frac{1}{2})^2} \quad \text{where} \quad n = 0, 1, 2 \ldots$$

(7.13)

Hence for $n = 0$, $E_{0}^{2D} = 4E_{0}^{3D}$ and the resulting 2-D exciton binding energy is of the order of 16 meV in quantum wells thinner than 50Å. Confinement does not affect the electron-phonon coupling which is primarily responsible for ionization of excitons into free electron-hole pairs. Hence, the increased binding energy in two dimensions produces strong excitonic contributions to the linear and non-linear optical properties even at room temperature. Excitonic features have been clearly observed in PL from high quality quantum wells at 300K [24,25] which is not the case in bulk GaAs. Detailed calculations [22,23] that take band mixing, image charges and effective mass mismatch in the well and barriers into account show that the exciton binding energies in very thin quantum wells can even exceed the limit predicted for the 2-D case of a simple hydrogen-like atom.

Excitons in 2 dimensions can also be treated like single particles instead of a many body system. An approximate exciton wave function in two dimension can be expressed in
terms of individual wave functions for an electron, a hole and their interaction. Thus

$$\Phi = \chi_e(\tau_e)\chi_h(\tau_h)\phi_{ex}(\rho)$$  \hspace{1cm} (7.14-a)

where

$$\phi_{ex}(\rho) = \sqrt{\frac{2}{\pi \lambda_{ex}}} \exp\left(-\frac{\rho}{\lambda_{ex}}\right)$$  \hspace{1cm} (7.14-b)

The exciton wave function $\phi_{ex}$ represents the relative in-plane motion of the 2-D electron and hole pair and $\lambda_{ex}$ is its radius. $\chi_e(\tau_e)$ and $\chi_h(\tau_h)$ are the electron and hole envelope wave functions describing confined quantum well states. The above expressions assume that a weak Coulomb interaction between the electron and hole does not perturb the confined system. The exciton radius $\lambda_{ex}$ is determined variationally so as to maximize the binding energy given by [26,27]

$$E^{2D}_b = -\frac{\hbar^2}{2m^* \lambda_{ex}^2} + \frac{e^2}{4\pi e} \langle \Phi \left| \frac{1}{\rho} \right| \Phi \rangle$$  \hspace{1cm} (7.15)

Figure 7.5 shows the dependence of the exciton radius and binding energy on the well width in quantum wells of different materials. The radius shrinks with $L_z$ and then increases below a critical value. This is because the electron and hole wave functions get increasingly localized with shrinking well width. However, below a critical width the energy levels get pushed to the top of the well and the wave functions spill into the barrier material. This causes a delocalization of the exciton and a subsequent increase in its radius. The binding energy shows the opposite trend. As $L_z$ decreases, the exciton binding energy increases from its 3-D value in the well medium to the theoretical 2-D limit (~ 4 times the 3-D value) Once the wave functions start spilling into the barrier, the exciton becomes bulk-like character and its binding energy approaches the 3-D value of the
barrier material. The II-VI quantum well excitons exhibit larger binding energies and smaller radii than the III-V ones due to the greater polarity (ionicity) of the former.

4 Exciton Lifetimes in Quantum Wells

Excitons in quantum wells are stable eigenstates of the well and have relatively long lifetimes as compared to the uncorrelated electron-hole pairs. One of the first theoretical investigations of the relationship between the spectral linewidth of exciton luminescence and their radiative lifetimes was done by Feldmann et al [28]. Ideally, excitonic absorption and PL are direct band to band transitions with $\Delta k = 0$. However, in practice the excitonic linewidth is not $\delta$-like and possesses homogeneous and inhomogeneous contributions. In high quality wells where the interface and alloy disorder are low, the acoustic phonon interaction determines the spectral linewidth $\Delta(E)$ at low temperatures. The effective transition strength for the exciton is obtained by sharing the $k = 0$ oscillator strength (which corresponds to a $\delta$-like lineshape) equally among all the states within the spectral width $\Delta(E)$. Only the fraction of the excitons $r(E)$ that possess momenta within the $\Delta(E)$ linewidth contribute to recombination. Using a simple model of Maxwell Boltzmann statistics, the effective 2-D oscillator strength $F_x^{2D}$ is given by [28]

$$F_x^{2D} = f_0 E_B^{2D} \frac{m^*}{(m_e + m_h)} \frac{r(E)}{\Delta(E)} \quad (7.16-a)$$

where

$$r(E) = \frac{\int^\Delta(E) D(E) f(E) dE}{\int^\infty D(E) f(E) dE} = 1 - \exp\left(-\frac{\Delta(E)}{k_B T}\right) \quad (7.16-b)$$

where $m^*$ is the reduced mass of the electron and hole, $f_0$ is the dipole matrix element connecting Bloch states in the valence and conduction bands, $D(E)$ is the 2-D density of
states which is a constant for free particles and $k_B$ is the Boltzmann constant. The relationship between the oscillator strength and radiative lifetime $\tau$ is given by [29]

$$\tau \propto \frac{1}{F_x^{2D}} \quad \text{or} \quad \tau \propto \frac{1}{E_B^{3D}} \left( \frac{m_e + m_h}{m^*} \right) \frac{\Delta(E)}{r(E)}$$  \hspace{1cm} (7.17)

Figure 7.6 shows the dependence of exciton lifetime on well width and temperature. The increase in lifetime with increasing well width is in part due to the reduction in binding energy as the well width increases. If the increase in lifetime is purely due to reduction in the binding energy, then $\tau$ should increase by a factor of 4 from a true 2-D exciton when $L_z \rightarrow 0$ to a true 3-D exciton for large $L_z$. However, this has not been observed in most quantum wells studied. An accurate quantitative description of the exciton lifetime can only be obtained by including the real band structure in the calculation and in particular, the variation of the valence band structure with $L_z$. The lower graph in figure 7.6 shows the exciton lifetime vs temperature. When the exciton linewidth $\Delta(E) \ll k_B T$ then $\tau \propto T$. On the other hand, if $\Delta(E) \gg k_B T$ then $\tau \propto \Delta(E)$. In a typical quantum well, the influence of $\Delta(E)$ on $\tau$ is difficult to quantify because there may be a number of inhomogeneous factors contributing to the linewidth [30,31]. Thus, a variation of radiative lifetime with temperature is only predicted for free excitons because of the variations in $\Delta(E)/r(E)$. 
Figure 7.1: Band structure of a direct bandgap III-V semiconductor in the vicinity of the zone center ($k \approx 0$). $\Gamma_6$ represents the conduction band and $\Gamma_8$ ($\Gamma_7$) are the valence (spin-orbit split-off) bands (reference [10], pg. 42).

Figure 7.2: Absorption spectrum of bulk GaAs at different temperatures. The Stokes shift in the absorption edge with increasing temperature is due to the shrinkage in the bandgap of GaAs with temperature. The spike in the absorption edge is due to excitonic contributions (reference [7], pg. 58).
Figure 7.3: GaAs/AlGaAs quantum well together with energy band diagram. Energy levels in the valence and conduction bands are quantized due to confinement ($hh =$ heavy hole band, $lh =$ light hole band).
Figure 7.4: 1. Absorption spectra at 2K from a series of quantum wells. The thinner wells show a series of peaks that are excitonic contributions from various quantized levels in the wells. 2. Well widths plotted versus exciton energies (reference [2]).
Figure 7.5: 1. Exciton radius versus well width. 2. Binding energy versus well width (reference [27]).
Figure 7.6: 1. Experimental exciton decay time versus well width. 2. Decay time versus temperature (reference [28]).
References


CHAPTER VIII

Exciton Dynamics in Growth Interrupted GaAs Quantum Wells

1 Introduction

An important issue associated with the growth of ultra-thin quantum wells (qw) is structure related disorder that occurs at the interfaces. This atomic level disorder, also known as interface roughness, can significantly modify the electronic and optical properties of thin quantum wells. Interface disorder can be reduced by interrupting the epitaxial growth at each interface [1,2]. Roughness that occurred on an atomic scale during continuous growth smoothens into large islands hundreds of angstroms in diameter, when growth is interrupted upon changing from one layer species to the next. In GaAs/AlGaAs quantum wells, these islands give rise to well regions that vary in thickness by one or more atomic monolayers (1 ML = a/2 or 2.83Å) and the excitons sample distinct well widths due to the large island size (>> exciton Bohr radius $a_0$). This is evident as fine structure in the excitonic photoluminescence (PL) where two or more peaks are observed for example in transitions associated with the ground heavy hole state [3-8]. While these studies have mainly focused on the cw emissions associated with excitons localized to regions of different well widths, there have been few detailed studies of their dynamics. Such studies are valuable as they provide information on island sizes, effects of inter-island
exciton diffusion, as well as the character of excitons, i.e. free versus bound, that may arise due to the nature and extent of interface roughness.

One of the first calculations on the effect of temperature on exciton lifetimes in quantum wells was performed by Feldmann and co-workers [9]. It was shown that lifetimes ($\tau$) of free excitons exhibit a linear dependence on temperature while those localized at defects or impurities show no dependence on temperature (7). Hence it should therefore be possible to determine the free or bound nature of excitons localized at monolayer well width fluctuations, by studying $\tau$ as a function of $T$.

One time resolved study reported that exciton lifetimes in N monolayer (ML) regions of a quantum well were much shorter (100 ps) than those localized at N+1 monolayer regions (200 ps) [10]. It was proposed that excitons created in the N ML well regions had two decay paths available; i.e. they may diffuse within their lifetimes to N+1 ML regions of the well or recombine directly. However, excitons at the N+1 ML region are localized by a step potential and hence can only recombine directly. Thus, exciton lifetimes are strongly modified by their diffusion. No detailed temperature dependence of the exciton lifetimes was reported in this study [10]. Another time resolved PL experiment in single GaAs quantum wells that showed exciton transfer from an N $\rightarrow$ N+1 ML well region due to diffusion was reported by Damen et al [11]. The measured exciton diffusion time from a N $\rightarrow$ N+1 monolayer region was 250 ps. This study also showed that diffusion effects strongly modify exciton lifetimes in the N and N+1 ML wide well regions.
Bergmann et al studied the lifetimes of free and bound excitons in acceptor doped single GaAs quantum wells [12]. The bound excitons had almost the same lifetimes as the free excitons of the well and increased with $T$ thus contradicting predictions in [9] which predicted that localized exciton lifetimes should exhibit no temperature dependence. It was proposed [12] that the observed $r$ vs $T$ behavior of the bound excitons was due to thermal interaction with the free excitons, thus causing a delocalization of the bound exciton momentum states and giving rise to a temperature dependence on lifetime.

How does interaction between excitons localized at different quantum well regions that vary in width by one monolayer (due to interface fluctuations) affect their lifetimes? In this study we examine such effects in undoped GaAs/AlGaAs single quantum wells by tuning the lattice temperature. Since the diffusion length can be varied through temperature, the latter provides a means to systematically study the effects of diffusion and interfacial island size on exciton lifetimes in regions of different well width.

We direct attention to PL from the $n = 1$, heavy hole ($hh$) exciton in undoped ultra-thin (18Å to 30Å wide) GaAs/AlGaAs single quantum wells grown by molecular beam epitaxy methods, using growth interruption. PL emissions from the wells show clearly resolved doublets whose energy separations are consistent with a single monolayer fluctuation in well width. The high energy peaks corresponds to PL from the $N$ monolayer regions of the wells and the lower ones to PL from the $N+1$ monolayer regions. We shall refer to the high and low energy peaks as the $E_x^N$ and $E_x^{N+1}$ excitons respectively.
The temperature dependence of the cw PL shows exciton oscillator intensity switching from the low energy $E_{x}^{N+1}$ state to the higher energy $E_{x}^{N}$ state as $T$ increases. Complementary time resolved PL studies reveal that both excitons are characterized by essentially the same lifetime $\tau$ at a given temperature. This temporal behavior indicates the absence of inter-island coupling through diffusion. This result is unlike that reported in references [10,11] where the lifetimes of the $E_{x}^{N}$ and $E_{x}^{N+1}$ excitons were very different. Our data (similar to [12]) does not follow predictions in [9] related to the temperature dependence of $\tau$ for localized excitons. We reconcile our results with [9-11] in terms of relevant length scales on which the exciton in-plane momentum is conserved.

2 Experiment

Two samples containing several single GaAs/Al$_{0.32}$Ga$_{0.68}$As quantum wells of varying widths were grown on undoped (001) GaAs substrates using Molecular Beam Epitaxy (MBE) methods. The barrier thickness was 300Å so that the individual wells were isolated from one another. The first sample (II-62) containing wells of widths 7, 11, 18, 28, 39 and 80 MLs, was grown at a substrate temperature of 640°C with a 90s growth interrupt at the inverted (GaAs on AlGaAs) interface. The second sample (II-61) containing wells of widths 5, 9, 14, 25, and 46 MLs, was grown at a substrate temperature of 590°C with a 90s growth interrupt at both quantum well interfaces. The well widths were measured by counting the reflection high energy electron diffraction (RHEED) intensity oscillations in-
situ during growth. In order to precisely control the quantum well (qw) width by monitoring the RHEED intensity, the substrate was not rotated during growth. Taking into account that non-uniformities occur during growth without rotation, this work was done using samples from regions of the wafer near where the RHEED was performed.

CW experiments were performed using the 6471Å Kr ion laser line for excitation. Signals were analyzed using a Raman spectrometer with 0.25 meV resolution. Time resolved PL measurements were performed using a cavity dumped Rhodamine 6-G dye laser source. The dye emission tuned to 2.03 eV had a pulse width of 5 ps and a 1 MHz repetition rate. PL decays were analyzed by a low temporal dispersion monochromator with 1-2 meV spectral resolution which adequately resolved the $E_x^N$ and $E_x^{N+1}$ excitons in all the wells. The signals were detected using a microchannel plate photomultiplier tube with 42 ps transit spread time. The resulting full width at half maximum (FWHM) of the instrument response was 47 ps. Exciton lifetimes were determined with an accuracy of 5 ps after deconvolving the instrument response. All measurements were performed in a continuous flow liquid helium cryostat over a temperature range of 5-120 K.

3 Results and Discussion

3.1 Continuous Wave Experiments

Figure 8.1 shows the low temperature (5K) cw PL measured using a low excitation density of less than 10 W/cm$^2$ from the two samples. No significant variation in lineshape was observed as the intensity was varied between 0.1 to 10 W/ cm$^2$. The top curve shows
PL spectra from wells in sample II-62 that was grown at a relatively high substrate temperature of 640°C with growth interrupted at the inverted interface. These spectra are similar to those in figures 10 and 18 in reference [6]. The higher substrate temperature increases the mobility of the deposited atomic species leading to smoother interfaces. The three thinnest wells (7/8 MLs, 11/12 MLs, 18/19 MLs) in this sample show clearly resolved doublets that appear as a weak high energy shoulder and a strong low energy peak with energy separations of 19, 13 and 7 meV respectively. The thicker 28 and 39 ML wells do not exhibit resolvable fine structure in their excitonic PL. The weak high energy shoulder on the 80 ML well that appears 3.4 meV above the main peak is due to luminescence associated with the light hole \((lh)\). The lower curve shows PL spectra from wells in sample II-61 (grown at a lower substrate temperature of 590°C and growth interrupted at both interfaces) where atomic mobilities are lower that hence produce rougher interfaces. PL from these wells show no fine structure and exhibit considerable statistical broadening consistent with greater interface roughness. The above results were verified by repeating measurements at different spots on the sample.

Figure 8.2.1 shows the energy separation between excitons \(E_x^N\) and \(E_x^{N+1}\) in the 7, 11 and 18 ML qws of sample II-62. The solid line is a calculation of the confinement energy for electrons and holes in a finite barrier square well using GaAs and AlGaAs parameters [13]. The good agreement with data indicates that the PL fine structure is most likely due to monolayer fluctuation in well widths.
Photoluminescence excitation (PLE) spectra from the 7, 11 and 18 ML wells confirm that the PL doublets are not due to the light hole exciton with measured $hh$-$lh$ separations for these wells being about 40, 32 and 24 meV respectively [14]. We also considered impurities as a source of the observed PL doublets. Acceptor impurities were ruled out as possible candidates on account of their high binding energies [15]. Donor impurities in GaAs have typical exciton binding energies of the order of 5-6 meV and could therefore be possible causes [15-17]. We argue this possibility to be unlikely based on our time resolved exciton lifetime studies which reveal that the excitons in the doublets have nearly the same lifetimes. Previous results have however showed that impurity bound excitons have characteristically different lifetimes than free excitons in a quantum well [10,12]. Moreover, impurity contributions should also be evident in the thicker 28 and 39 ML wells which was not observed.

The inset in figure 8.2.1 shows schematically the normal and inverted interfaces of a GaAs/AlGaAs quantum well that give rise to two intrinsic well widths, $L_z$ and $L_z + \Delta L$ ($\Delta L = 1$ ML). The normal interface is smoother than the inverted interface because the high quality GaAs surface permits the AlGaAs layer to spread smoothly. Hence this interface forms islands with diameters much larger than the exciton Bohr radius ($d >> a_0$) [6]. The inverted interface on the other hand is rougher due to the poorer AlGaAs surface quality. Thus fluctuations at this interface occur on an atomic scale and the excitons perceive an average roughness at this interface as shown by the dashed line in the inset. Therefore, excitons confined to these wells sample two distinct widths arising from ML
fluctuations at the normal interface which accounts for the doublets in the PL spectra [10]. Figure 8.2.2 summarizes the temperature dependence of the $E_x^N$ and $E_x^{N+1}$ luminescence peak energies for the 7, 11 and 18 ML wells of sample II-62.

Figures 8.3.1-3 show normalized cw PL spectra from the 7, 11 and 18 ML wells respectively of sample II-62 as a function of temperature along with theoretical fits that are discussed below. In the thicker 11 and 18 ML wells at low $T$, $E_x^N$ appears as a weak high energy shoulder whereas the intensity of $E_x^{N+1}$ is strong. As $T$ rises, the $E_x^N$ to $E_x^{N+1}$ intensity ratio become comparable around 65K. Above 100K, the $E_x^{N+1}$ emission is essentially quenched and the PL is almost entirely due to $E_x^N$. In contrast, the $E_x^{N+1}$ luminescence from the 7 ML well is not quenched even at 120K (possibly due to the larger ML energy separation of 19 meV). The PL associated with $E_x^{N+1}$ and $E_x^N$ fit well to Gaussian curves weighted by thermal occupancy factors and amplitudes chosen as fitting parameters. A Gaussian lineshape is an adequate description when broadening is primarily controlled by interface fluctuations which is true for our samples [18].

3.2 Time Resolved Experiments

Radiative lifetimes of the $E_x^{N+1}$ and $E_x^N$ excitons in the II-62 sample quantum wells were determined via time resolved PL spectroscopy. The aims were two-fold: 1. To understand the switching in cw PL intensity from $E_x^{N+1}$ to $E_x^N$ as temperature rises. Bounds on the interfacial island sizes can be deduced once the radiative lifetimes are known 2. To
determine whether $E_x^{N+1}$ excitons exhibit the same linear dependence of $\tau$ on $T$ predicted for free excitons and if the lifetimes of $E_x^{N+1}$ and $E_x^N$ differ due to confinement [9].

Figure 8.4 shows the PL decay intensity vs time on a semi-log scale for the 11 ML well. Spectra from the other wells of II-62 were of a similar nature. The dots are data and solid lines are fits to mono-exponential obtained after deconvolving the instrument response. Figures 8.5.1 and 8.5.2 shows the PL lifetimes which increases almost linearly with temperature up to 60 K, for the $E_x^{N+1}$ and $E_x^N$ excitons in the 7 and 11 ML wells in sample II-62. The slightly larger values of the $E_x^{N+1}$ exciton lifetimes over $E_x^N$ is consistent with the weak direct dependence of $\tau$ on well width $L_z$ [19]. The lifetimes merge at high temperatures (figures 8.5.1 and 8.5.2) because the $E_x^{N+1}$ excitons get thermally delocalized from their confining potential and subsequently transform into $E_x^N$ excitons. The flattening of $\tau$ vs $T$ in the 7 and 5 ML wells (figures 8.5.1, 8.5.3) beyond 65 K indicates the onset of non-radiative processes that diminish radiative lifetimes [20].

Exciton lifetimes increase from about 225 ps at 5K to 400 ps at 90K. The low temperature values of $\tau$ agree well with published results [9] but the high temperature exciton lifetimes in the II-62 sample are much lower than those reported in [9] (1.5 ns). The exciton lifetimes in the 5 and 9 ML quantum well of the II-61 sample (figure 8.5.3) that was grown at a lower substrate temperature exhibit a similar increase with temperature as in [9]. This difference in the slopes of the $\tau$ vs $T$ graphs of II-62 and II-61 though not fully understood, are consistent with previous reports [6].
In reference [9] it was shown that the exciton lifetime $\tau$ depends upon the quantity \[ \frac{\Delta(T)}{1 - e^{-\frac{kT}{\Delta(T)}}} \] where $\Delta(T)$ is the exciton spectral linewidth around wave-vector $k = 0$. In the limit $\Delta(T) \ll k_0 T$, $\tau \propto k_0 T$ while if $\Delta(T) \gg k_0 T$, $\tau \propto \Delta(T)$. At 5K, the observed exciton linewidths in our quantum wells is $\Delta(T) \sim 10$ meV and therefore the latter condition is more applicable. Our cw PL measurements confirm that the linewidth increases approximately linearly with temperature which is consistent with the nearly linear $\tau$ vs $T$ behavior in our samples. We attribute the variation of exciton linewidth with temperature to acoustic phonon contributions [8].

In contrast to reports in [10,11], diffusion effects are not evident in our low temperature data because the $E_x^N$ and $E_x^{N+1}$ excitons have nearly the same lifetimes and exhibit the same $\tau$ vs $T$ behavior. In what follows we trace these characteristics to the large interfacial island sizes in our quantum well samples. Diffusion effects strongly reduce the lifetime of $E_x^N$ excitons by providing another decay path (i.e. diffusion from $N \rightarrow N+1$ ML well region to become an $E_x^{N+1}$ exciton) in addition to direct annihilation. This process can occur at low temperature only if the low temperature exciton diffusion length ($L_D$) is larger than the island diameter ($d$). In this case, it has been previously interpreted that within their radiative lifetimes, $E_x^N$ excitons diffuse to the thicker well regions and thermalize into $E_x^{N+1}$ states [4,6,10,11]. The reverse process is inhibited at low temperatures by the ML confining potential of the island. If we consider the opposite
condition where the low temperature exciton diffusion length is smaller than the island
diameters (i.e. $L_D < d$), then excitons created at N ML regions cannot diffuse to N+1 ML
regions within their radiative lifetimes. The resulting lifetimes of $E_x^N$ and $E_x^{N+1}$ excitons are
independent and would differ only slightly due to a weak dependence of $\tau$ on $L_z$.

Our time resolved data indicates that decay times of excitons in N and N+1 ML
islands are not coupled through diffusion at low temperatures. Hence, the low temperature
integrated cw $E_x^{N+1}$ to $E_x^N$ PL intensities measure the relative areas of the N+1:N ML
regions in the well as sampled by the laser beam. Since the diffusion length $L_D$ increases
with temperature, for appropriate island sizes, the $E_x^{N+1}$ excitons could diffuse to the N
ML regions at higher temperatures thus contributing to the observed switching in the cw
PL intensity from $E_x^{N+1}$ to $E_x^N$ at high temperatures.

The switching of the cw PL oscillator strength from $E_x^{N+1}$ to $E_x^N$ with temperature
deserves further discussion. In the narrowest 7 ML well, the integrated $E_x^N$ oscillator
strength increases with $T$ but remains less than that of $E_x^{N+1}$ even at 125K. However a
complete depletion of the $E_x^{N+1}$ states in the thicker 11 and 18 ML wells is observed. This
implies that exciton motion from N+1 $\rightarrow$ N ML regions is preferred over the reverse (N
$\rightarrow$ N+1) process in the wider well. The former (N+1 $\rightarrow$ N) involves overcoming the ML
barrier potential which is aided by thermal energy as the sample is heated. Why does the
reverse process (N $\rightarrow$ N+1 motion) which is not inhibited by an energy barrier not occur?

One possible explanation is that when an $E_x^N$ exciton migrates to an N+1 ML
region, it must expend it's excess (ML) energy before thermalizing in the $E_x^{N+1}$ state. At
low temperatures (<100K), acoustic phonon emission is the most likely energy loss mechanism. It is possible that an $E_x^N$ exciton is unable to diffuse to an N+1 ML region and thermalize there within it’s radiative lifetime, via acoustic phonon emission. Calculations of energy loss rates through acoustic phonon emission range from 0.01 meV/ps to 0.1 meV/ps depending on the underlying assumptions of the calculations [21,22]. If the energy loss rate lies closer to the lower predicted value, then an $E_x^N$ exciton would be unable to thermalize into an $E_x^{N+1}$ state within it’s radiative lifetime through this channel. This may explain the observed cw PL intensity switching from $E_x^{N+1}$ to $E_x^N$ in the thicker wells. The absence of a complete switch in the $E_x^{N+1}$ to $E_x^N$ intensity of the 7 ML well is likely due to the larger monolayer separation energy (19 meV) and/or possibly a smaller areal density of the N ML well regions that are saturated prior to the depletion of the N+1 ML states. Results revealing intensity switching similar to ours have also been observed in other works but have not been addressed in detail [4, 6 and references therein]. For example, in figure 4 of reference [4] the intensity of the $E_x^{N+1}$ peak at 2K is slightly larger than that of the $E_x^N$ peak in a 50Å/50Å wide GaAs/AlGaAs superlattice sample. As the temperature increases the oscillator intensity shifts to $E_x^N$ and by 70K the $E_x^{N+1}$ peak is almost completely quenched. Similar results were also obtained in multiple quantum well samples [4]. These results have been interpreted as being due to “thermalization effects”.

When charge neutrality is retained (ambipolar limit) the diffusion length is given by

$$L_D = \left( \frac{\mu k_B T \tau}{e} \right)$$

where $\mu$ is the carrier mobility [23]. Using $\mu = 1500 \text{ cm}^2/\text{Vs}$ [11] and the
measured values of $\tau$ from figure 8.5, we estimate that $L_D$ increases from about 0.2\mu m to 1.0\mu m as $T$ increase from 10K to 120K. The values of $L_D$ hence place limits on the island sizes, which in sample II-62 must be greater than the low temperature diffusion length of 0.2\mu m. However, the onset of switching in the cw PL intensities above 50K implies that the spatially separate N and N+1 ML thick regions of the well are now coupled via diffusion. We hence estimate the island sizes to be about 0.4-0.5\mu m. Our estimated low temperature diffusion length (0.2\mu m) agrees with the results of Damen et al where inter-island exciton transfer was observed [11].

The last issue we address is why the excitons $E^N_x$ and $E^{N+1}_x$ display the same $\tau$ vs $T$ behavior? Feldmann et al predicted that the exciton lifetime depends on the spectral linewidth $\Delta(T)$ around $k = 0$ [9]. Excitons bound to impurities or defects have highly localized momentum states with a spectral linewidth that is independent of $T$. Hence, lifetimes of bound excitons are expected to show little or no temperature dependence [9]. Experiments on doped GaAs quantum wells however showed that both free and bound excitons displayed the same dependence of $\tau$ on $T$ [12]. In our wells (sample II-62), there is a randomly varying potential on the \mu m length scale arising from island formation during growth interruption. This breakdown of in-plane translational invariance could lead to exciton localization [7]. However, the in-plane potential perceived by excitons on the length scale of the exciton Bohr radius ($a_0 \sim 100\text{Å}$) is still uniform as the island diameters ($d \sim 0.4-0.5$ \mu m) are much larger than $a_0$. For this reason, the $E^{N+1}_x$ excitons in our
samples are essentially free excitons of a N+1 ML well and hence exhibit the same \( \tau \) vs \( T \) behavior as the \( E_x^N \) excitons.

4 Summary

We have probed exciton PL using cw and time resolved techniques in growth interrupted quantum wells. Fine structure in the cw PL is attributed to transitions from N and N+1 ML thick regions in the well formed during growth interruption. In the cw experiments, the switching of oscillator strength from \( E_x^{N+1} \rightarrow E_x^N \) at high temperatures is attributed to the diffusion of localized carriers from thicker well regions to narrower well segments before recombination. Time resolved studies reveal that the excitons \( E_x^N \) and \( E_x^{N+1} \) have nearly the same lifetimes for all \( T \) indicating the absence of exciton diffusion from \( N \rightarrow N+1 \) ML islands at low temperatures. Hence the low temperature ratio of the \( E_x^{N+1} / E_x^N \) integrated cw PL intensity provides a measure of the relative areas of N+1 to N ML regions sampled by the laser beam. The time resolved and cw results together place limits on the island sizes, \( d \sim 0.4-0.5 \mu m \). Since \( d \gg a_b \), the \( E_x^{N+1} \) excitons perceive a uniform in-plane potential on a length scale larger than the exciton Bohr radius. A consequence of this effect is the identical \( \tau \) vs \( T \) behavior for both excitons in the quantum wells.
Figure 8.1: Continuous wave PL from two quantum well samples at 5K for low excitation density. The numbers alongside the peaks indicates the monolayer (ML) well widths. The top curve shows the PL from sample II-62 that was grown at a higher substrate temperature of 640°C. The PL linewidths here are narrow due to interface smoothening and the thinnest wells show a fine structure. Wells that exhibit ML fluctuations in widths are indicated with two well widths; ex. 7-8 ML etc. Arrows indicate the position of the high energy shoulder due to the $E_N^*$ exciton in the 7, 11 and 18 ML wells. The high energy shoulder on the 80 ML well is due to the light hole exciton. The bottom curve is for sample II-61 that was grown at a lower substrate temperature of 590°C. The PL linewidths from wells in II-61 show considerable broadening arising from interface roughness.
Figure 8.2: 1. Calculated confinement energy for holes and electrons in a finite square well using material parameters for GaAs. The hollow dots are the monolayer energy separation measured for the 7-8, 11-12 and 18-19 ML wells of sample II-62 while the solid line is a calculation. The inset shows a schematic diagram of monolayer fluctuations in well width at the normal interface in growth interrupted quantum wells. The PL doublet arises from excitons confined in regions of the well that vary in thickness by 1 monolayer. 2. Photoluminescence energy of the $E_x^N$ and $E_x^{N+1}$ excitons vs temperature. The PL energies track the bandgap of GaAs as it shrinks with $T$. 

1 ML = 2.83Å
Figure 8.3: Normalized (with respect to peak intensity) continuous wave PL from the 7, 11 and 18 ML wells as a function of temperature. Dots are data while solid lines are fits to two gaussians weighted by thermal occupancy factors. The $E_x^{N+1}$ exciton peak is strong at low temperatures while the $E_x^N$ exciton oscillator becomes stronger as $T$ increases. The high energy tails at high temperatures arise from band to band free carrier recombination.
Figure 8.4: Exciton PL decay as a function of time for 11 ML well on a semi-logarithmic plot. The scatter in the profiles that appear towards the end of the decay are due to detector noise. Dots are data while solid lines are fits to mono-exponentials over a range of 3 decades change in the PL intensity. The $E_x^{N+1}$ exciton lifetimes are slightly higher than the $E_x^N$ exciton lifetimes.
Figure 8.5: $E_x^N$ and $E_x^{N+1}$ exciton lifetimes as a function of temperature in 1. The 7 ML well of sample II-62 2. The 11 ML well of sample II-62 and 3. 5 and 9 ML wells of sample II-61.
References


CHAPTER IX

Exciton Lifetimes at High Carrier Densities in GaAs Quantum Wells

1 Introduction

The dynamics of exciton formation and relaxation in GaAs quantum well heterostructures have been extensively studied using ultrafast spectroscopy techniques [1-4]. These studies are important because quantum confinement enhances the excitonic binding energy which in turn influence nonlinear optical properties of GaAs heterostructure devices even at room temperature. Devices such as quantum well lasers operate at high current densities (carrier density \( n_0 \sim 10^{12} \, \text{cm}^{-2} \)) where many body effects become important. A rough estimate of the critical (Mott) density where a semiconductor \( \rightarrow \text{metal} \) transition occurs is given by the criterion \( n_0 a_B^2 \sim 1 \) [5,6], where \( a_B \) is the exciton Bohr radius. In GaAs quantum wells \( a_B \sim 100 \, \text{Å} \), which gives a critical density \( n_0 \sim 10^{12} \, \text{cm}^{-2} \). Beyond the Mott density excitons are screened by the free carriers and are no longer stable bound states. Much of the recent work on exciton dynamics has focused on determining the role of temperature \( (T) \) and excitation wavelength on excitonic PL rise times following picosecond or femtosecond excitation. Shah et al [1] studied the PL rise time as a function of \( T \). The slow PL rise time at low \( T \) was interpreted in terms of the long time required for large wave-vector excitons to thermalize at low temperature, in order for
them to recombine radiatively. PL rise time studies under resonant conditions revealed a long heavy-hole \((hh)\) PL rise time when excitation coincided with the light hole \((lh)\) transition [2,3]. Resonant excitation studies also revealed dephasing effects in these systems [4]. These studies and others [7], focused on understanding PL rise time at low carrier concentrations as a function of temperature.

This report presents preliminary results on time resolved PL from GaAs quantum wells at high carrier densities, a study not attempted previously quite possibly due to complications in the analysis arising from many body effects. The PL decay lineshape of the \(n = 1, \ hh\) exciton in very thin (15Å - 30Å wide) single GaAs/AlGaAs quantum are monitored as a function of carrier concentration \(n_0\) and sample temperature \(T\). At low \(n_0\) \((10^8-10^{10} \text{ cm}^{-2})\), the lifetimes exhibit the usual linear dependence on \(T\) and the decays yield excellent fits to mono-exponentials with randomly varying residuals. At intermediate \(n_0\) \((10^{10}-10^{11} \text{ cm}^{-2})\), the PL emissions show strong deviations from a mono-exponential behavior. They exhibit a convex profile when observed on a logarithmic scale which suggests a cooling process. When \(T\) is increased, the convex shaped luminescence profile becomes mono-exponential as in the low density regime. At peak \(n_0\) achieved in our experiments \((10^{12}-10^{13} \text{ cm}^{-2})\) an initial mono-exponential is observed. The lifetime of this component is independent of \(T\) over 5K-75K. The mechanisms underlying the above observations are discussed by studying the influence of temperatures on various stages of the photoluminescence.
2 Experiment

Two high quality GaAs/Al$_{0.32}$Ga$_{0.68}$As quantum well samples were fabricated at GTE laboratories using MBE techniques on undoped (001) GaAs substrates. The first sample (designated II-61) had 5 quantum wells of thickness 5, 9, 14, 24 and 46 monolayers (ML) (14Å and 24Å respectively). The second sample designated (UCSB-A) had two wells of thickness 6 and 9 ML. The AlGaAs barrier was 300Å thick ensuring that all wells were uncoupled. The well widths were monitored in situ by counting the RHEED oscillations. Low temperature time resolved PL measurements were conducted to investigate the dynamics of the $n = 1$, hh exciton decay in the quantum wells using the same setup described in chapter VIII. Experiments were performed using excitation energies above (2.03 eV) and below (1.9 eV) the barrier bandgap to ensure that the results were not due to charge transport effects in the AlGaAs barrier. Decays from only the thinnest wells of II-61 (5 ML and 9 ML) were studied in detail because of instrument limitations. Results from the 9 ML well of II-61 were confirmed by repeating measurements on the 9 ML well of UCSB-A. In order to achieve high carrier densities in a controlled manner, the incident laser beam was expanded to 5 mm in diameter and directed onto a high quality achromatic doublet lens (mounted on a precision translational stage) that focused light on the sample. The carrier density on the sample was varied through the diameter of the focused laser spot by adjusting the position of the translational stage. By this simple technique, we achieved controlled carrier density variations over three orders of magnitude.
3 Results and Discussion

Figure 9.1 shows the cw PL at 20K from the 5 and 9 ML wells of sample II-61 and the 9 ML well of sample UCSB-A. The PL emissions are inhomogeneously broadened from interface roughness. Impurities are absent in the spectral region of interest. PL from the 9 ML wells of the two samples appears at approximately the same energy and have nearly the same linewidths indicating that the wells are of similar quality.

Figure 9.2.1 shows typical low carrier density \((n_0 = 2 \times 10^9 \text{ cm}^{-2})\), \(n = 1\), hh exciton PL decays on a semi-log scale from three quantum wells at 10 K. The dotted lines are data while the solid lines are fits to mono-exponential after deconvolving the instrument response. The top graph in figure 9.2. also shows the instrument response with a FWHM of 50 ps. Figure 9.3 shows the PL decay lifetime \(\tau\) vs \(T\) for the wells in figure 9.2 using the same \(n_0\) as before. The PL lifetimes at low densities scale linearly with temperature as discussed in chapter VIII [7]. The saturation of \(\tau\) for the 5 ML around 55 K and its subsequent decrease is due to the prominence of non-radiative decay channels in thinner quantum wells [8]. Finally, the excitonic \(\tau\) vs \(T\) behavior from the two 9 ML wells are nearly identical.

Figure 9.4 shows PL at 10K decays from the above wells at a high carrier concentration \(n_0 \sim 10^{12} \text{ cm}^{-2}\). The decays plotted on a semi-log scale show strong deviations from the mono-exponential profiles seen previously at low densities. This structure in the PL, though evident in the thinnest 5 ML, well is more pronounced in the 9
ML wells. We examine the effects of temperature on this complicated decay pattern focusing attention on the II-61, 9 ML well as results from the other wells are similar. The above experiments when repeated for excitation below the barrier bandgap yielded similar results albeit with lower absolute PL intensities.

3.1 PL vs \( n_0 \) and \( T \)

Figures 9.5.1-9.5.3 show the heavy hole PL decay on a semi-log scale from the II-61 9 ML quantum well for a series of temperatures 10K, 40K and 85K respectively and \( n_0 \) ranging from \( 10^9 \) to \( 10^{12} \) cm\(^2\). The graphs across increase in \( T \) while those down increase in \( n_0 \). The first row of decays correspond to a low carrier density of \( 2 \times 10^9 \) cm\(^2\) similar to figure 9.2. When \( n_0 \) is increased to \( 5 \times 10^{10} \) cm\(^2\), the decay acquires a convex profile (second row of figures) suggesting that the PL lifetime changes continuously during the decay. The convexity of the decay is clear at 10K. It is not as prominent at 40K (dotted line corresponding to a mono-exponential shown for comparison) and practically non-existent at 85K. Hence, the convex decay profile is clearly affected by the sample temperature. A mono-exponential function does not fit this decay even at 85K where it visually appears mono-exponential in nature. Note that the convex lineshape becomes mono-exponential towards the end of the luminescence. When \( n_0 \) is increased from \( 5 \times 10^{10} \) cm\(^2\) to \( 5 \times 10^{11} \) cm\(^2\) the convexity gets more pronounced and prolonged.

The last row of graphs show the PL intensity vs \( T \) at \( n_0 \sim 10^{12} \) cm\(^2\). An initial mono-exponential decay not previously observed, appears at this density. This is followed
by the convex profile decay (observed at intermediate \( n_0 \)) which finally evolves into a pure mono-exponential near the end of the decay. Thus, the PL decay occurs in three stages at very high carrier densities.

Figure 9.6 shows a typical decay at the highest carrier density in our experiments (\( n_0 = 3 \times 10^{12} \, \text{cm}^{-2} \)) for a series of temperatures. The dots are data and the solid lines are fits using three single exponentials, one for each stage of the decay. At 5K, the first and third stages fit well to mono-exponentials whereas the convex shaped second stage (\( \tau \) changes continuously with time) yields a poor fit to a mono-exponential. A correction to a mono-exponential is a function of the form \( \exp(-t/(\tau_0 + \alpha t)) \) where \( \tau_0 \) signifies the lifetime at the onset of stage 2 and \( \alpha \) represents the cooling rate. These fits however yield nearly the same value \( \alpha \) over a wide range of temperatures. Since \( \alpha \) represents a cooling rate, it should be proportional to \( (T_{\text{ex}} - T_{\text{l}}) \) where \( T_{\text{ex}} \) is an effective exciton temperature at the onset of this stage and \( T_{\text{l}} \) is the lattice (sample) temperature. Thus \( \alpha \) should reduce with increasing sample temperature. The insensitivity of \( \alpha \) to \( T \) is most likely due to the small intensity range (factor of 2 to 3) available to fit the data from the second stage. In comparison, lifetimes for the first and third stages are extracted by fitting mono-exponentials to data over a range of one order of magnitude while \( \tau \)'s at low densities are obtained by fitting to data over a range of more than 3 orders of magnitude.

The results of the piece-wise mono-exponential fits are summarized in figure 9.7. The lifetime of stage 1 remains at 525 ps till 40K and then reduces to 420 ps at 85K. The
lifetime of the second stage is very long at 5K (1.4 ns) where the convexity is most pronounced and reduces to 800 ps at 85K and is now almost equal to the corresponding \( \tau \) of stage 3 (770 ps). Thus, the lifetime of the initial mono-exponential is relatively insensitive to temperature while the cooling (2nd) segment tends towards a mono-exponential and becomes contiguous with the third stage as \( T \) rises. The third stage yields good fits to single exponentials over all temperatures and the resulting \( \tau \) rise linearly with \( T \) as in the low carrier density case (figure 9.3).

3.2 Discussion

Eccleston et al [3] observed fast initial decays at the onset of PL while studying the \( hh \) exciton rise times using resonant excitation coincident with the \( lh \) energy. The carrier density in their study was \( 1.5 \times 10^{10} \) cm\(^2\) and the measured \( \tau \)'s that were of the order of 50 ps, reduced rapidly with increasing temperature. These results were interpreted in terms of heating of cold excitons by LA phonons. In our work, the initial mono-exponential appears only when \( n_0 \sim 10^{12} \) cm\(^2\). The lifetime of this component is relatively insensitive to the temperature and the decay times measured for this component are an order of magnitude larger than in [3]. All these differences suggest that the phenomenon observed here is not due to phonon induced heating of excitons.

Our experiment were performed using 5 ps pulses at 2.03 eV and a 1 MHz repetition rate. The PL produced by one pulse decays completely before the next one arrives and we therefore eliminate photon buildup effects as a possible explanation for our
observations [9]. We also rule out the light hole emission as a candidate because the $hh$-$lh$ separation which is of the order of 36 meV in the 9 ML well is clearly resolved by the spectrometer. Hence we do not expect any $lh$ signatures when the spectrometer is tuned to the $hh$ emission. Thus, we conclude that the origin of this emission is intrinsic to the $n = 1$, $hh$ transition and most likely due to high carrier density effects.

At high carrier concentrations, electrons excited to various momentum states in the conduction band scatter primarily via electron-electron interaction to the $k = 0$ state [9,10]. LO phonon scattering is weak at low temperatures because of the low phonon occupation number. LO phonon scattering is also suppressed by screening at high carrier concentration ($n_0 = 10^{12} - 10^{13} \text{ cm}^{-2}$) [11,12]. Carrier induced screening should also inhibit formation of excitonic bound states. A rough estimate of the critical density when excitons are completely screened is given by $n_0 a_B^2 \sim 1$, where $a_B$ is the exciton Bohr radius [5,6]. This estimate also gives the density at which many body effects should become significant. In our experiments we have $a_B \sim 100\text{Å}, n_0 > 10^{12} \text{ cm}^{-2}$, and hence the above relationship is easily satisfied. Thus the initial mono-exponential decay in our data for the highest $n$ case might be due to PL from electron-hole recombination rather than of excitonic origin. If this were indeed the case, the PL should be blue shifted by the exciton binding energy ($\sim 16$ meV) when the excitons dissociate into free electron-hole pairs. This effect was not observed within instrument resolution (2-3 meV). However, it is possible that this blue shift from the exciton dissociation might be compensated by a red shift from bandgap shrinkage that is expected at high carrier densities [5]. It is worth noting that in cw PL
studies a very slight red shift (2-3 meV) was observed in the PL emission energy when the carrier density was increased by about 5 orders of magnitude starting from a nominal sheet density of $10^8$ cm$^2$. This small bandgap shrinkage observed at the highest $n_0$ in the cw experiments would have gone undetected in the time resolved experiments within the spectral resolution of our monochromator.

We propose the following argument for the origin of the convex decay profile, starting from the initial mono-exponential. The convex shaped decay occurs for $n_0$ over a range $2 \times 10^{10}$ to $5 \times 10^{11}$ cm$^2$. The initial mono-exponential has a lifetime $\tau \sim 500$ ps and lasts 600 ps and then gives way into a convex shaped decay. Thus if at $t = 0$, $n_0 = 10^{12}$ cm$^2$, it should reduce to $3 \times 10^{11}$ cm$^2$ by the onset of the second stage ($t = 600$ ps) from recombination alone. The resulting carrier density now lies in the range where the convex decay shape is most prominent. As the density reduces, screening also decreases and weakly bound excitons can form. These excitons get heated by the electron-hole plasma which transfers some kinetic energy to the excitons during collision, and then recombines radiatively. At low temperatures, this hot exciton gas is far from thermal equilibrium with the lattice. It cools to the lattice temperature as it decays thus giving rise to a convex decay profile. As $T$ increases, the temperature difference between the exciton gas at the onset of cooling and the lattice temperature reduces and hence the cooling effects are not evident at high lattice temperatures.

At 5K, the exciton lifetime at the onset of the second stage is very long (1.4 ns, figure 9.7). The effective temperature corresponding to this lifetime obtained by
extrapolating the low powered $\tau$ vs $T$ data (figure 9.3) is 130K. This temperature is a measure of the average excitonic kinetic energy ($\sim 11$ meV) at the end of the first stage of the PL decay. The excitons cool to an equilibrium lifetime of 400 ps (third stage value, figure 9.7) at 5K. The corresponding lifetimes at low carrier density at 5K obtained from figure 9.3 is 260 ps. The difference of 140 ps probably arises from the local heating of the sample by the tightly focused laser beam.

Other investigators may have observed slightly convex PL decay profiles due to high carrier density effects and not attributed it to the above reason [13]. In this paper, the authors note that the PL decay time exhibits a nonlinear (nearly logarithmic) dependence on $T$ and they also detect the presence of a weak secondary component with a long lifetime. A similar effect was reported in another study also [14]. In our experiments, we observe the onset of non-exponential decay profiles for $n_0 \sim 2 \times 10^{10}$ cm$^{-2}$. The decays here can be fitted to the sum of two exponential where the weaker component has a longer lifetime. The use of a single exponential to fit the data yields non-random residues that exhibit an oscillatory behavior. The effective lifetimes obtained by forcing an exponential function to fit such decays resulted in a sublinear dependence of the PL lifetimes on temperature similar to the results reported in [12].
4 Summary

We have studied the effects of very high carrier density on the PL decay in thin GaAs quantum wells. At low $n_0$ ($10^8$-$10^{10}$ cm$^2$) the usual linear $\tau$ vs $T$ behavior is observed. At intermediate $n_0$ ($10^{10}$-$10^{11}$ cm$^2$), the PL becomes convex shaped and exhibits cooling effects. When $n_0 > 10^{12}$ cm$^2$, the decay occurs in three stages. A mono-exponential component is observed at the onset. This stage followed by the convex shaped decay evident at intermediate densities and it finally evolves into a single exponential as in the low density case. The lifetime of the initial mono-exponential is insensitive to $T$ whereas the convex profile gets washed out as the sample temperature rises. The value of $n_0$ at the end of the first stage is about $3 \times 10^{11}$ cm$^2$ which falls in regime where the convex decay profiles are observed. The final stage of the 3 step decay yields good fits to exponential functions and show a linear $\tau$ vs $T$ relationship as in the low density case. Simple piece-wise mono-exponential fits illustrate the qualitative features of the multi-stage decay discussed above.

We attribute the convex shaped decay at intermediate densities to cooling of excitons. Hence this process is sensitive to $T$, and vanishes at high temperatures when the thermal energy of the excitons is close to the lattice temperature. The origin of the initial mono-exponential component seen at high $n_0$ is unclear. Given that $n_0a_0^2 \sim 1$ (Mott density regime) in our experiments and excitons are most likely screened, one likely explanation is that the initial mono-exponential arises from band to band electron-hole
recombination. No blue shift was observed in the PL at high densities (due to the
dissociation of excitons) to substantiate this hypothesis. However, this may be due to
compensation by bandgap shrinkage [5,6]. Valuable information on the PL decay
dynamics can be obtained from a knowledge of the Fermi levels as a function of time. In
order to extract this information, integrated energy spectra must be measured at various
time points after excitation (say, 0.1, 1, 10, 100, 500, 1000 and 2000 ps). This peak in the
integrated (energy) PL intensity will give a measure of the Fermi surface and hence the
instantaneous carrier density. Any measurable shift in the peak PL emission energy with
time will also provide insight into effects of screening and bandgap shrinkage.

This work has reported preliminary results on PL at high densities. Although a
number of issues remain unanswered, it points clearly to the need for more experimental
and theoretical effort in this fascinating and complex field.
Figure 9.1: CW PL from the n=1, hh excitons in the 5 and 9 ML wells from the II-61 sample at 20K. The narrow PL linewidths (9 and 11 meV for the 5 and 9 ML wells respectively) indicates the high quality of the quantum wells. The inset in the figure shows the PL another 9 ML well (UCSB-A) grown separately. The emission features are similar to those of II-61.
Figure 9.2: Time resolved PL emissions from the 3 quantum wells at a low carrier density ($n_0 = 2 \times 10^9 \text{ cm}^{-2}$). The PL emissions are mono-exponential. The data points in the figure are dots and exponential fits to the data are solid lines. The top graph in the figure also shows the IRF with a FWHM of 50 ps.
Figure 9.3: \( \tau \) vs \( T \) for the wells from the two samples. The lines in the plots are guides to the eye. A linear behavior is exhibited between 15 to 80 K for the 9 ML wells. The exciton lifetime starts dropping after 55K in the 5 ML well as the non-radiative contribution to the decay becomes significant.
Figures 9.4: Time resolved PL from 3 wells at very high carrier density ($n_0 = 3 \times 10^{12}$ cm$^{-2}$). The PL decay occurs in several stages. The multi-stage decay is more prominent in the 9 ML wells than in the 5 ML one.
Figure 9.5: Decays for II-61 9 ML well at 1. 10K, 2. 40K and 3. 85 K. Each individual figure shows the PL decays for three carrier densities. At the lowest carrier densities (a), the decays are all mono-exponential. At intermediate carrier densities (b), the PL decay appears convex shaped on a semi-log scale which suggests a continuously changing lifetime. At peak carrier densities, (c), a mono-exponential component appears. This is then followed by the convex decay profile as in the intermediate density case and finally the decay ends as a mono-exponential.
Figure 9.6: Three PL decays for the 9 ML well at peak carrier densities for a series of temperatures. The solid lines are 3 piece-wise mono-exponentials fits, one for each stage.
Figure 9.7: Lifetimes obtained from single exponential fits for the three stages of the PL decay at peak carrier density. The lifetime of the initial stage is relatively insensitive to temperature whereas the lifetimes of the second and third stages fuse together as sample temperature is raised. This is consistent with the observations of figure 9.5 where the convex shaped stage straightens into a mono-exponential.
References


CHAPTER X

Future Work

This thesis studied the elastic properties and magnetoelastic effects in body centered cubic (bcc) cobalt (Co). The measured elastic constant provide useful inputs to compare the results of first principle calculations with. For completeness and as a further test of both theory and experiment, the elastic constants of the face centered cubic (fcc) structure of cobalt should also be measured. FCC cobalt samples have recently been stabilized on MgO and sapphire substrates to thickness' of the order of 1000Å and work is underway in our laboratory to complete these measurements. Another metal that has been stabilized in various phases is nickel (Ni). Elastic constants of various phases of Ni will also provide a critical test of theoretical calculations.

Magnetoelastic effects are very prominent in thin (< 100Å thick) magnetic films. They are also manifest the spin wave dispersion in the form of large uniaxial anisotropies in bcc Co films that are over 200Å thick. Magnetic anisotropies are extremely sensitive to growth conditions. Hence a systematic study that carefully correlates anisotropy effects to film thickness and growth conditions is highly desirable from the technological point of view. This will facilitate fabrication of well characterized films with similar properties for device applications.
Magnetoelastic effects can also give rise to interactions between spin waves and phonons that can be observed using Brillouin spectroscopy [1]. Garnets are excellent candidates to study these effects using Brillouin spectroscopy. Compared to metals, they have relatively large optical penetration depth and hence provide large interaction volumes for phonon and magnon scattering. Second, the low saturation magnetization for these materials implies that the spin wave frequencies are usually quite low (lower than phonon frequencies). Hence the spin wave frequency can be swept through the phonon modes by tuning the magnetic field in order to study phonon-magnon interactions. The 3-\(d\) transition metals are not good candidates for such studies because the spin wave frequencies here are usually higher than the phonon frequencies even at zero magnetic field, due to the larger saturation magnetization in these materials.

Brillouin spectroscopy can also be extended to study plasmon excitations in semiconductor doped GaAs quantum wires. Plasmon dispersion has been studied in doped quantum wells using Raman spectroscopy but in 1-D quantum wire systems they have extremely low energies (0.1-1 meV) and cannot be probed by Raman techniques. These energies however lie at the high end of the Brillouin spectrum and can in principle be studied using Brillouin spectroscopy. However, these experiments will be demanding and have stringent requirements. The spectrometer must be accurately calibrated in order to operate at such a large free spectral range. Since the plasmon scattering cross section is enhanced by resonant excitation and quantum wires have their absorption band in the 700-
severe luminescence arising from the GaAs substrates on which the wires are patterned. On possible way to circumvent this problem is by patterning the wires on a ZnSe buffer instead of directly on GaAs. ZnSe is very closely lattice matched with GaAs and has its bandgap in the blue. Hence PL from the substrate will be reduced. There will be PL arising from the wire itself but that could be minimized using a prism or grating.

PL from GaAs quantum wells and wires must also be examined in detail using high carrier density excitation. Studies of the integrated (in energy space) PL emission at various time point after excitation will provide insight into the role of many body effects that are predicted to occur at high density.
References

APPENDIX A

Rayleigh Wave Dispersion in Anisotropic Supported Films

The Rayleigh wave dispersion along [1T0] is analyzed in detail for a (110) grown supported cubic film on a cubic substrate. The equation of motion in the absence of piezoelectric effects and external forces is given by

\[ \rho \frac{\partial^2 u_i}{\partial t^2} = \frac{\partial T_{ij}}{\partial x_j} \quad (i, j, l = 1, 2, 3) \]  (A.1)

where \( \rho \) is the density of the medium, \( T_{ij} \)'s are elements of the stress tensor. Hooke's law provides a relation between stress \( T \) and the strain \( S \)

\[ T_{ij} = C_{ijkl} S_{kl} \]  (A.2)

In the linear regime, the strain field \( S \) is related to the displacement fields \( u_i \)'s by

\[ S_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \]  (A.3)

The equation of motion can be rewritten using equations (A.2) and (A.3) as

\[ \rho \frac{\partial^2 u_i}{\partial t^2} = C_{ijkl} \frac{\partial^2 u_k}{\partial x_j \partial x_l} \quad (i, j, k, l = 1, 2, 3) \]  (A.4)
In the Voigt notation, the elasticity tensor is described by two indices instead of the usual four using the scheme 11 → 1, 22 → 2, 33 → 3, 23, 32 → 4, 13, 31 → 5, 12, 21 → 6. The fourth rank elasticity tensor, \( C \), reduces to a 6x6 matrix after taking all the symmetry properties of the elasticity tensor into account. The above notation can be extended to the stress and strain tensors also and we obtain 6x1 vectorial representations for them. In a cubic solid there are 3 independent elastic constants, \( c_{11}, c_{12} \) and \( c_{44} \). When the geometric axes coincide with the normal crystal axes, the form of the elasticity tensor is

\[
C = \begin{pmatrix}
  c_{11} & c_{12} & c_{12} & 0 & 0 & 0 \\
  c_{12} & c_{11} & c_{12} & 0 & 0 & 0 \\
  c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\
  0 & 0 & 0 & c_{44} & 0 & 0 \\
  0 & 0 & 0 & 0 & c_{44} & 0 \\
  0 & 0 & 0 & 0 & 0 & c_{44}
\end{pmatrix}
\] (A.5)

Sometimes the geometric axis has to be rotated to obtain convenient solutions to the wave equation. The elasticity tensor must then be transformed into this co-ordinate system. The transformation properties of the elasticity tensor are given in [A.1] and the transformed elasticity tensor \( C' \) is given by

\[
C' = MCM^T
\] (A.6)

Figure A.1 shows a film of bcc Co grown on the (110) plane of GaAs. The \( xy \) axes are the normal geometric axes and the primed \( x'y' \) axes are the body transformed axes obtained by a 45° counter-clockwise rotation about the \( z \) axis. Thus \( z = z' \). The transformation matrix \( M \), for the above rotation is given by
Hence the transformed elasticity tensor becomes

\[
M = \begin{pmatrix}
0.5 & 0.5 & 0 & 0 & 0 & 1 \\
0.5 & 0.5 & 0 & 0 & 0 & -1 \\
0 & 0 & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 0.717 & -0.717 & 0 \\
0 & 0 & 0 & 0.717 & 0.717 & 0 \\
-0.5 & 0.5 & 0 & 0 & 0 & 0
\end{pmatrix}
\]

(A.7)

Hence the transformed elasticity tensor becomes

\[
C' = \begin{pmatrix}
a_{11} & a_{12} & c_{12} & 0 & 0 & 0 \\
a_{12} & a_{11} & c_{12} & 0 & 0 & 0 \\
c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & c_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & c_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & a_{66}
\end{pmatrix}
\]

(A.8)

where

\[
a_{11} = \frac{c_{11} + c_{12} + c_{44}}{2} \\
a_{12} = \frac{c_{11} + c_{12} - c_{44}}{2} \\
a_{66} = \frac{c_{11} - c_{12}}{2}
\]

In the primed frame, the free surface of the film is at \( x' = -h \) and the substrate extends towards \( x' \to \infty \). The principal crystal directions \([1\overline{1}0]\) and \([001]\), lie along the \( y' \) and \( z' \) respectively.

A Rayleigh wave propagating along \( y' \) (primes dropped in future) that decays into the bulk (\( x \) direction) is given by

\[
u_i = u_{i0} \exp(i k b x) \exp[i k (y - v t)] \quad (i = x, y, z)
\]

(A.9)
where the quantity $b$ (eigenvalue, same as $l_i$ used in chapter 3) is in general a complex quantity and $u_0$ is the amplitude of partial wave. In the Voigt notation, the stress and strain tensors assume the form

$$T_{ij} = C_{ij} S_j \quad (i,j = 1,6) \quad (A.10)$$

After substituting (A.9) into (A.2) and (A.3) we get

$$
\begin{pmatrix}
T_1 \\
T_2 \\
T_3 \\
T_4 \\
T_5 \\
T_6
\end{pmatrix}
= i k
\begin{pmatrix}
a_{11} b u_x + a_{12} u_y \\
a_{12} b u_x + a_{11} u_y \\
c_{12} (b u_x + u_y) \\
c_{44} u_z \\
c_{44} b u_z \\
a_{66} (u_x + b u_y)
\end{pmatrix}
\quad (A.11)
$$

The wave equation in the Voigt notation takes the form

$$\nabla_i T_j = \rho \frac{\partial^2 u_i}{\partial t^2} \quad (i = 1,3, J = 1,6) \quad (A.12)$$

From equations (A.11) and (A.12) we get

$$
\begin{pmatrix}
(a_{11} b^2 + a_{66} - \rho v^2) & (a_{12} + a_{66}) b & 0 \\
(a_{12} + a_{66}) b & (a_{66} b^2 + a_{11} - \rho v^2) & 0 \\
0 & 0 & (c_{44} b^2 + c_{44} - \rho v^2)
\end{pmatrix}
\begin{pmatrix}
u_x \\
u_y \\
u_z
\end{pmatrix}
= 0 \quad (A.13)
$$

For solutions to exist to the $u_i$'s the determinant of the above matrix equation must be zero. The determinant factors into fourth and second order equations in $b$ where the former equation corresponds to the Rayleigh mode. The eigenvalue equation in $b$ is solved
numerically. Solutions in the film are a superposition of four partial waves while those in the substrate are composed of two partial waves:

\[
\begin{align*}
    u_f &= \sum_{n=1}^{4} f^{(n)}(\alpha_{f(n)}) \exp(i k b_f n x) \exp[i k (y - vt)] \\
    u_s &= \sum_{n=1}^{2} f^{(n)}(\alpha_{s(n)}) \exp(i k b_s n x) \exp[i k (y - vt)]
\end{align*}
\]  
(A.14)

The superscripts \(f\) and \(s\) refer to the film and substrate respectively. \(\alpha_f(n)\)'s are eigenvectors constructed using the eigenvalues \(b(n)\)'s, and \(f(n)\)'s are arbitrary complex constants corresponding to the partial wave amplitudes. Solutions to the eigenvectors are obtained by substituting the above forms of \(u_f\) into equation (A.12). If we consider only the Rayleigh mode solution we can neglect the in-plane \(u_z\) component. This simplifies (A.13) into a 2x2 equation

\[
\begin{pmatrix}
(a_{11} b^2 + a_{66} - \rho v^2) & (a_{12} + a_{66}) b \\
(a_{12} + a_{66}) b & (a_{66} b^2 + a_{11} - \rho v^2)
\end{pmatrix}
\begin{pmatrix}
\alpha_x \\
\alpha_y
\end{pmatrix} = 0 \quad (A.15)
\]

which can be solved for the eigenvectors.

The above machinery is used to solve for the Rayleigh wave velocity for the film-substrate combine as a function of \(k\). For a given wave-vector \(k\), an arbitrary value of velocity is chosen to solve equation (A.13) for the eigenvalues \(b(n)\)'s in the film and substrate. Next, the eigenvectors are computed in the two media. The value of velocity must be such that the mechanical boundary conditions at the interfaces are satisfied.

1. Continuity of displacement at \(x = 0\).

\[
\sum_{n=1}^{4} u_f^{(i)} = \sum_{n=1}^{2} u_s^{(i)} \quad (i = x, y)
\]
2. Continuity of stress at $x = 0$.

\[
T_{1s} = T_{1f} \\
T_{6s} = T_{6f}
\]

3. The free surface of the film is stress free, i.e.

\[
T_{1s} = T_{1f} \\
T_{6s} = T_{6f}
\]

at $x = -h$. The above boundary conditions give rise to homogeneous set of six equations that may be written in the form

\[
\sum_{s=1}^{6} M_{if} f^{(n)} = 0 \tag{A.16}
\]

Solutions to exist for the amplitudes $f(n)$'s only if the determinant $|M| = 0$. The lowest value of velocity that satisfies (A.16) is the Rayleigh wave solution. It is computed in an iterative manner by starting with a low trial value of velocity and increasing it till equation (A.16) is satisfied. In order to extract the elastic constants of a thin film using Rayleigh wave dispersion, the calculated dispersion is fitted to the experimental data treating the elastic constants as fitting parameters.
Figure A.1: Top view of (110) plane bcc Co film on a GaAs substrate. The primed axes are obtained by a $45^\circ$ counter clockwise rotation about $z$. In this frame, the principal crystal directions [1 1 0] and [0 0 1] lie along the $y'$ and $z'$ respectively. The free surface of the film is at $x' = -h$ and the substrate grows in the direction $x' \to \infty$. 
APPENDIX B

Spin Wave Dispersion in BCC Cobalt

The procedure for calculating the spin wave dispersion is outlined for a (001) grown film where magnetization is not parallel to the applied external field. Expressions for the components of \( \frac{dm}{dt} \) given in equation 6.8 are used as starting points for the derivation. The transformation for the wave-vector components is similar to that of the magnetization components. In the primed co-ordinate system, the applied field is along \( z' \) and the wave-vector propagates perpendicular to this field. Hence \( q_z = 0 \). As a result we have \( q_x = q_x' \cos \phi \) and \( q_z = -q_x' \sin \phi \). The equation of motion for the time and spatially varying magnetization, \( m(t) \), is solved together with Maxwell’s equations of magneto-statics.

\[
\nabla \times h = 0 \quad \text{(B.1)}
\]

\[
\nabla \cdot (\mu + 4\pi m) = 0 \quad \text{(B.2)}
\]

\[
\frac{1}{\gamma} \frac{\partial M}{\partial t} = M \times \left[ H - \frac{1}{M} \nabla M E_{\text{anh}} + \left( \frac{2A}{M} \right) \nabla^2 M \right] \quad \text{(B.3)}
\]

Since \( \nabla \times h = 0 \), we can write \( h = -\nabla \Psi \) where \( \Psi \) is a scalar potential. The form of the scalar potential is a plane wave within the film and an exponentially decaying wave outside the film.
Substituting expressions for $\Psi$ into equations B.1-3, we obtain a homogeneous set of equations that may be cast in the form

$$
\begin{pmatrix}
4\pi iq_x \cos \phi & 4\pi iq_y & -q^2 \\
\frac{i\omega}{\gamma} & -H_x & iMq_y \\
H_y & \frac{i\omega}{\gamma} & iMq_y
\end{pmatrix}
\begin{pmatrix}
m_x \\
m_y \\
\Psi
\end{pmatrix} = 0
$$

(B.4)

where $q^2 = q'^2$. The terms $H_x$ and $H_y$ contain information on all the anisotropy fields and are given by

$$
H_x = \lambda_x + Dq^2 \text{ where } \lambda_x = H\cos(\phi_H - \phi) + \frac{2K_1}{M} (1 - \frac{1}{2}\sin^2 2\phi) + \frac{2K_U}{M} \cos^2 \phi + \frac{2K_P}{M}
$$

$$
H_y = \lambda_y + Dq^2 \text{ where } \lambda_y = H\cos(\phi_H - \phi) + \frac{2K_1}{M} (1 - 2\sin^2 2\phi) + \frac{2K_U}{M} (1 - 2\sin^2 \phi)
$$

(B.5)

The forms of $\lambda_x$ and $\lambda_y$ are obtained from equations 6.4 and 6.5. The homogeneous set of equations (B.4) possess solutions only if corresponding determinant vanishes which gives rise to an eigenvalue equation in $q$ given by

$$
q^6 + \frac{[\lambda_x + \lambda_y + 4\pi M]}{D} q^4 + \frac{\lambda_x \lambda_y + 4\pi M\lambda_y - \frac{\omega^2}{\gamma^2}}{D^2} - \frac{4\pi Mq_x^2 \sin^2 \phi}{D} \gamma^2 + \frac{(4\pi Mq_x^2)(\lambda_x \cos^2 \phi - \lambda_y)}{D^2} = 0
$$

(B.6)
The above equation gives rise to six solutions in $q_y$. The eigen-vectors $m_x$ and $m_y$ are constructed using equation B.4.

$$m_x' = u_x'h_x$$ and $$m_y' = u_y'h_x'$$

where

$$u_x' = M\left(\frac{H_x - \frac{i\omega}{\gamma} \frac{q_x}{(q_y)_x}}{H_xH_y - \frac{\omega^2}{\gamma^2}}\right)$$

and

$$u_y' = M\left(\frac{H_y - \frac{q_y}{(q_y)_y} + \frac{i\omega}{\gamma}}{H_xH_y - \frac{\omega^2}{\gamma^2}}\right)$$

(B.7)

$i = 1..6$ within the film and $i = 1$ outside the film (at $y < 0$ and $y > h$). The $h_y$ term has been eliminated through the relation $h_y = -\frac{q_x}{q_y}h_x$.

The magnetization $m^i$ is computed in terms of unknown amplitudes $h^i$s. The fields within and outside the film must satisfy the electromagnetic and exchange boundary conditions. These are given by the continuity of normal components of $B$ and tangential components of $H$ at $y = 0$, $h$ and $\frac{\partial m}{\partial n} = 0$ $y = 0$, $h$ respectively, where is $n$ is the outward normal to the film surface [1,2]. The boundary conditions give rise to a set of 8 equations in the amplitudes $h^i$s that can be reduced to 6 independent equations through linear combinations. This homogeneous set of equations gives rise to a 6x6 boundary condition determinant which must be zero for solutions to exist for the $h^i$s.

Surface like solutions are obtained when the eigenvalues $q_y$s are imaginary. These waves have a maximum amplitude near the surface and produce strong scattering of light.
Bulk-like solutions occur when the eigenvalues are real. These are standing wave modes and take values \((n\pi/h)\) where \(h\) is the film thickness. The exchange, gyromagnetic ratios and anisotropies are unknown parameters of the theory. The boundary condition determinant is employed in an iterative scheme where the least squared error between the calculated and measured spin wave frequencies is minimized with respect to the unknown parameters.
APPENDIX C

Magnetoelasticity Effects in BCC Cobalt

The forms of the elastic and magnetoelastic tensors together with expressions for the free energy are given here. The magneto-elastic tensor has the same form as the elastic tensor. For the (110) film, the form of the elasticity tensor is the same as in appendix A. The form of the free energy neglecting shear strains is given by

\[ F(e) = \frac{1}{2} \sum_{i,j} e_i c_{ij} e_j \ (i,j = x, y, z). \]

Hence

\[ F(e) = \frac{1}{2} [a_{11}(e_x^2 + e_y^2) + c_{11}e_z^2 + 2a_{12}e_x e_y + 2c_{12}(e_y e_z + e_z e_x)] \]  \hspace{1cm} (C.1)

where the \( a_i \)'s are defined in appendix A. The form of the elasticity tensor in the above coordinate frame for the (001) film is

\[
C = \begin{pmatrix}
c_{11} & c_{12} & c_{12} & 0 & 0 & 0 \\
c_{12} & c_{11} & c_{12} & 0 & 0 & 0 \\
c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & c_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & c_{44} & 0 \\
0 & 0 & 0 & 0 & 0 & c_{44}
\end{pmatrix}
\]  \hspace{1cm} (C.2)

The magnetoelastic tensor for this film has the same form as above. Neglecting shear strains the form of the free energy is

\[ F(e) = \frac{1}{2} [a_{11}(e_x^2 + e_y^2 + e_z^2) + 2a_{12}e_x e_y + 2c_{12}(e_y e_z + e_z e_x)] \]  \hspace{1cm} (C.3)
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