Magnetic Anisotropy, Spin Pinning and Exchange Constants of (Ga,Mn)As films

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We present a detailed investigation of exchange-dominated nonpropagating spin-wave modes in a series of 100 nm Ga$_{1-x}$Mn$_x$As films with Mn concentrations $x$ ranging from 0.02 to 0.08. The angular and Mn concentration dependences of spin wave resonance modes have been studied for both as-grown and annealed samples. Our results indicate that the magnetic anisotropy terms of Ga$_{1-x}$Mn$_x$As depend on the Mn concentration $x$, but are also strongly affected by sample growth conditions; moreover, the magnetic anisotropy of Ga$_{1-x}$Mn$_x$As films is found to be clearly linked to the Curie temperature. The spin wave resonance spectra consist of a series of well resolved standing spin-wave modes. The observed mode patterns are consistent with the Portis volume-inhomogeneity model, in which a spatially nonuniform anisotropy field acts on the Mn spins. The analysis of these exchange-dominated spin wave modes, including their angular dependences, allows us to establish the exchange stiffness constants for Ga$_{1-x}$Mn$_x$As films.

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I. INTRODUCTION

Incorporating Mn ions into III-V semiconductors makes it possible to achieve ferromagnetic order in semiconductor nanostructures. It is known that the spin waves in a magnetic system are determined by (and can be used to obtain) the exchange and magnetic anisotropy parameters, and are thus particularly useful for a quantitative understanding of ferromagnetism in Ga$_{1-x}$Mn$_x$As. In this paper we use ferromagnetic resonance (FMR), a powerful tool for investigating magnetic anisotropy and exchange constants in ferromagnetic materials, to investigate a series of Ga$_{1-x}$Mn$_x$As films with different Mn concentrations $x$. In all samples studied we observe a multi-mode spin wave resonance (SWR) spectrum. We will focus on the detailed description of such SWRs, and the relationship between these spin wave modes and the Mn concentration $x$ in Ga$_{1-x}$Mn$_x$As.

II. SAMPLE FABRICATION

A series of Ga$_{1-x}$Mn$_x$As films were grown by molecular beam epitaxy (MBE) on semi-insulating GaAs (001) substrates. A 100 nm-thick GaAs buffer was first grown at the substrate temperature 600°C to achieve an atomically flat surface. The substrates were then cooled to 250°C for growth of a 2 nm low temperature GaAs buffer, followed by 100 nm Ga$_{1-x}$Mn$_x$As layers with various Mn concentrations ($x = 0.02, 0.025, 0.04, 0.045, 0.055, 0.06, 0.07$ and $0.08$). The thickness of (Ga,Mn)As samples is specifically selected to reliably determine the effect of annealing on SWRs. All as-grown specimens exhibited ferromagnetic order, with Curie temperature $T_C$ ranging from 50 K to 80 K. Pieces of samples cleaved from each specimen were then annealed in N$_2$ gas for one hour at 280°C in order to examine the effect of annealing on magnetic properties. As a result of annealing $T_C$ of the samples increased to a range from 55 K to 115 K. Note that $T_C$ follows a general rule for both as-grown and annealed samples: it increases with increasing concentration $x$.

III. EXPERIMENTAL SETUP

In this study we have measured the angular dependence of FMR for each specimen (both as-grown and annealed) in three geometries. FMR measurements were carried out at 9.46 GHz using a Bruker electron paramagnetic resonance spectrometer. The applied dc magnetic field $H$ was in the horizontal plane, while the microwave magnetic field was acting vertically on the sample. The sample was placed in a suprasil tube inserted in a liquid helium continuous flow cryostat, which could achieve temperatures down to 4.0 K.

The Ga$_{1-x}$Mn$_x$As layers were cleaved into three square pieces with edges along the [110] and [100] directions. Each square piece was then placed in three different orientations. Geometry 1 is when the sample plane and the [110] edge are vertical, which allows measurement with dc magnetic field $H$ oriented at any angle between $H||[001]$ (normal orientation) and $H||[110]$ (in plane orientation). Geometry 2 is when the sample plane and the [010] direction are vertical, which allows us to measure FMR with field orientations between $H||[001]$ and the in-plane orientation $H||[100]$. Geometry 3 is when the sample plane is horizontal, allowing us to map out the FMR when $H$ is confined to the layer plane.

IV. RESULTS AND DISCUSSIONS

A. Magnetic Anisotropy

Multi-mode SWR spectrum was observed in all samples. Our analysis of the data was carried out as follows. We first obtained the magnetic anisotropies and $g$-factors...
by fitting the angular dependence of the strongest resonance line to a theoretical uniform FMR model using a nonlinear least squares method. Our fitting results show that the magnitude of perpendicular uniaxial anisotropy $H_{2\perp}$ has a tendency to increase with Mn concentration $x$, while the in-plane cubic anisotropy $H_{4\parallel}$ decreases with $x$. However, the relation between the magnetic anisotropy terms $K_{\perp}$ and $K_{\parallel}$ is strongly influenced by the individual growth condition of each sample. Furthermore, we find that the term $4\pi M_{eff} = 4\pi M - H_{2\perp}$ is linearly increasing with the Curie temperature $T_C$ in both as-grown and annealed specimens, and $H_{4\parallel}$ is monotonically decreasing with $T_C$, with the exception of annealed specimens with the highest values of $T_C$. The results are shown in Fig. 1. Since an earlier report shows an empirical relationship $T_C \sim p^{1/3}$, we immediately note that there exists a tight relationship between magnetic anisotropy and the hole concentration $p$. However, the data for as-grown and annealed samples follow different slopes as a function of $T_C$, suggesting that there exists a fundamental difference between as-grown and annealed samples regarding magnetic anisotropy. Nevertheless, the distinct characteristics which we observe may provide useful insights into the origin of magnetic anisotropy in Ga$_{1-x}$Mn$_x$As films.

B. SWR Spectra

It is the spin pinning at sample surfaces that induces spin-wave excitation in the FMR experiment. To investigate this feature in detail, we have chosen four specimens grown at similar conditions, with Mn concentration $x = 0.02, 0.04, 0.06$ and $0.07$. Figure 2 shows the SWR spectra at $T = 4$ K for both as-grown and annealed samples when the dc magnetic field $H$ is normal to the sample plane ($H||[001]$, i.e., $\theta_H = 0^\circ$). In both cases the SWR spectra consist of several well-resolved standing spin wave modes separated by roughly equal field increments. Note that the SWR fields are increasing with the Mn concentration $x$, except in the case of one annealed sample with Mn concentration $x = 0.07$. Importantly, the as-grown samples have a larger mode separation than the annealed samples, which indicates that the exchange stiffness constant $D$ is larger in the as-grown samples, as discussed later.

As $H$ is rotated away from the perpendicular orientation, the low-field spin wave modes gradually disappear, and eventually only one narrow resonance line remains at a critical angle $\theta_c$. We will refer to it as the uniform mode. For angle $\theta_H > \theta_c$ the spectrum generally consists of two wide resonance lines. We identify the mode lying at the higher field as an exchange-dominated surface spin wave mode. The angular dependence of the SWRs reveals the nature of surface spin pinning and its dependence on the magnetization orientation. Using the Puszkarski surface inhomogeneity (SI) model, it is found that, as the magnetization rotates from the perpendicular to the in-plane orientation, the surface spins are evolving from a strong pinning condition to a weak pinning condition, with a turning point at the critical...
angle $\theta_{\parallel,\perp}$. Note that for $\mathbf{H}||[001]$ the pinning is not only strong, but also nonlocalized, i.e., the magnetic spins appear to be affected by a spatially nonuniform anisotropy field\textsuperscript{[2]}, suggested by a linear mode separation at this orientation.

### C. Exchange Constants

We will now focus on the SWR spectrum obtained for $\mathbf{H}||[001]$. It is found that the positions of the SWR modes comply with a linear mode separation model for $\mathbf{H}||[001]$ when a symmetrical parabolic magnetic anisotropy is assumed along the growth direction $z$ ($|z| \leq L/2$): 

$$4\pi M^\ast(z) = 4\pi M^\ast(0)(1 - 4\varepsilon z^2/L)\textsuperscript{[2]}.$$  

Here $\varepsilon$ is the distortion parameter of the film, $L = 100 \text{ nm}$ is the film thickness, and $4\pi M^\ast(0) = 4\pi M - H_{2\perp} - H_{1\perp}$. In this situation the position of the $n$-th SWR mode for $\mathbf{H}||[001]$ is given by the Portis relation\textsuperscript{[2]}:

$$H_n = H_0 - (n - \frac{1}{2})(4/L)(4\pi M^\ast(0)\varepsilon)\frac{D}{g\mu_B}^{1/2}, \quad (1)$$  

where $\mu_B$ is the Bohr magneton, $n$ is an odd integer, and $H_0 = \omega/\gamma + 4\pi M^\ast(0)$ is the position of the theoretical uniform mode. Here $\omega$ is the angular frequency of the microwave field and $\gamma$ is the gyromagnetic ratio. The exchange stiffness constant $D$, which gives a measure of the strength of the exchange interaction, can be determined from the difference of the adjacent SWR modes by:

$$\frac{D}{g\mu_B} = \frac{\Delta H^2_{n1,n2}(z)^2}{16(n2 - n1)^2(4\pi M^\ast(0)\varepsilon)}. \quad (2)$$

Note that the value of $4\pi M^\ast(0)\varepsilon$, the depth of parabolic potential well, can be roughly evaluated from the field separation between the highest and the lowest SWR modes observed for $\mathbf{H}||[001]$. In the special case where only two modes are observed (see Fig. 2, $x = 0.07$), it is interesting that $\Delta H_{1,3} \approx 4\pi M^\ast(0)\varepsilon$, so that $D$ can be satisfactorily estimated simply from the separation of the two modes.

Using Eq. (2), we can obtain the value of $D/g\mu_B$ for all samples. The data for four optimally-grown samples are plotted as function of Mn concentration $x$ in Fig. 3. The figure clearly shows that $D$ is closely related to $x$. In particular, for as-grown samples $D$ decreases very quickly as $x$ increases. On the other hand, although the data for annealed samples suggest that $D$ also decreases as $x$ increases, the change is much smaller in this case. The most important characteristic shown in Fig. 3 is that $D$ decreases after annealing, especially for low Mn concentration samples. This experimental result is quite surprising, because $T_C$ – which usually go hand-in-hand with the strength of the exchange interaction – are higher in the annealed samples than in the as-grown films. One possible explanation is that the average distance between

![FIG. 3: The exchange stiffness constant $D$ as a function of Mn concentration $x$ for as-grown and annealed GaMnAs films. The lines are linear fits.](image)

### V. SUMMARY AND CONCLUSIONS

In summary, we carried out a detailed experimental study of SWRs in thin Ga$_{1-x}$Mn$_x$As films with a wide range of Mn concentrations $x$. The analysis of the data allowed us to establish the following important magnetic properties of these films: magnetic anisotropy, surface spin pinning, and the exchange stiffness constant. The magnetic parameters obtained from this analysis were studied as function of $x$ and/or $T_C$ of the specimens. We observe a significant change of magnetic properties between as-grown and annealed samples. The results clearly show that the study of SWRs provides valuable information about the strong correlation between structural and magnetic properties of these materials.

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