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Annealing effect on the magnetization reversal and Curie temperature in a GaMnAs layer

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Abstract:

Vibrating Sample Magnetometer measurements were performed on a ferromagnetic (Ga,Mn)As thin film. We report Curie temperature (T_c) up to 142 K in annealed 200 nm GaMnAs layer. This result is remarkable and comparable with the T_c obtained in GaMnAs for small thicknesses $t < 50$ nm. Our result reveals the high quality of the sample albeit the presence of 3 different chemical species, and a large thickness. After comparison with previous work obtained in GaMnAs, we show that for film of up to $t = 200$ nm the out diffusion based annealing process is not thickness limited.

Keywords: GaMnAs, Annealing, Carrier density and Curie temperature

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1. Introduction:

The discovery of ferromagnetism in dilute magnetic semi-conductors (III,Mn)V has made it possible to demonstrate a number of new principles of spintronic device operations [1,2]. Their exact mechanisms of the magnetic interaction are still under discussion. Ferromagnetism in (Ga,Mn)As layers is commonly related to magnetic exchange interaction between the conduction holes and localized magnetic moments of the Mn_{Ga} atoms (Mn atoms located in Ga sites) [3]. The ferromagnetic ordering temperature (T_c) of GaMnAs is directly related to the content of Mn and

the holes density. Several groups shows that GaMnAs layers with high Mn doping are not suited in the framework of the Zener mean field [4,5]. They prove that only a fraction of the Mn dopant is magnetically active. Indeed, due the limited solubility of Mn in bulk GaAs, the low temperature conditions necessary for epitaxial growth of GaMnAs, induces defects such as Mn interstitials and As antisites that act as compensating donors and hence suppress ferromagnetic ordering [6,7]. The Curie temperature in GaMnAs strongly depends on the growth, in particular on the amount of compensating defects, i.e., the Mn atoms on interstitial

positions Mn_I (close either to As or Ga sublattices) and As antisites on the Ga sublattice [8]. Various techniques have been proposed to increase T_c by careful annealing of MBE-prepared samples – it is possible to increase the T_c in GaMnAs [9]. Several factors act to favoring higher T_c . Indeed the growth parameters and post-growth annealing play a crucial role in limiting T_c . Wang et al. have studied the effect of these two parameters and they found a linear dependence of T_c and M_s with Mn moment density [10]. The highest Curie temperature up to 190 K was obtained for annealing temperatures T_a just below the growth temperature and with 10 % effective Mn concentration [11], while long annealing times and low annealing temperatures result an increase of T_c [12,13].

In this work, we have investigated the higher qualities of a 200 nm thick (Ga,Mn)As layer through a characterization by X-ray diffraction and VSM techniques. We reported the annealing effect on Curie temperature and we obtained a high T_c in spite of the large thickness for GaMnAs contrary to previous works.

2. Samples growth and experimental details:

The samples under study have been grown by low temperature molecular beam epitaxy (LTMBE). It has a structure consisting of a 200 nm layer of $Ga_{1-x}Mn_x$ As deposited on a GaAs (100) substrate. In a first approximation and without taking into account the exact location of Mn atoms (interstitial or substitutional sites), the Mn concentration was estimated by high resolution x-ray diffraction [14]. The (Ga,Mn)As layer was deposited at 713°C.

After the growth, the sample was thermally annealed under N_2 atmosphere at 200°C for 7 h. We have studied a GaMnAs sample, as grown (a) and annealed (b). The nominal Mn concentration was around $x=0.1$. We applied a magnetic field in the plane of the sample along [1-10], and varied it from -500 Oe to 500 Oe.

3. Results and discussion:

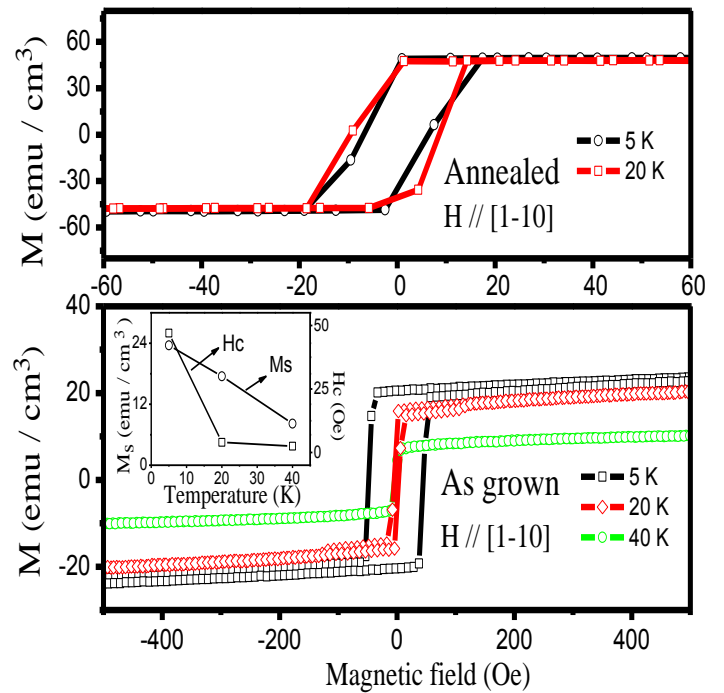


Fig.1. Magnetization curves as a function of the magnetic field along [1-10] for the as grown (a) and the annealed (b) GaMnAs layer. $T=5$ K (black line), $T=20$ K (red line), $T=40$ K (green line). The inset shows the dependence of both the saturation magnetization and the coercive field as a function of temperature for the As-grown GaMnAs layer.

Fig.1 presents the VSM data as a function of the magnetic field for the GaMnAs sample, both before and after annealing. The data shows a decrease of the saturation

magnetization (M_s) with increasing temperature. Indeed, for $T=5$ K, the M_s is in the vicinity of 23.6 emu/cm^3 and the hysteresis loop is square. At higher temperature, it loses this character, and the magnetization reversal occurs at lower field, $M_s \approx 17.5 \text{ emu/cm}^3$ at $T=20$ K and $M_s \approx 8 \text{ emu/cm}^3$ at $T=40$ K (Inset .Fig.1). This modification of M_s and the slope of hysteresis loops can be explained by a result from a competition in the plan of the layer between the cubic anisotropy ($K_{4\text{par}}$) according to $[100]$ and growth uniaxial anisotropy ($K_{4\text{perp}}$) according to $[110]$. In addition, at low temperature the cubic anisotropy dominates the uniaxial contribution. However, at higher temperature the latter becomes stronger [15-17].

We also noticed that the value of the coercive field (H_c) is fairly high at low temperature (Inset. Fig.1). It decreases abruptly to 45 Oe at $T=5$ K from 4 Oe and 2.5 respectively at $T=20$ K and 40 K. After annealing, we observe that the saturation magnetization increases abruptly to 48 emu/cm^3 for both $T=5$ K and 20 K. The difference between the shapes of the two hysteresis loops disappears. This is due to removal of antiferromagnetically coupled $\text{Mn}_{\text{Ga}}\text{-Mn}_{\text{I}}$ pairs, which does take part in the ferromagnetic phase in the as-grown sample [18].

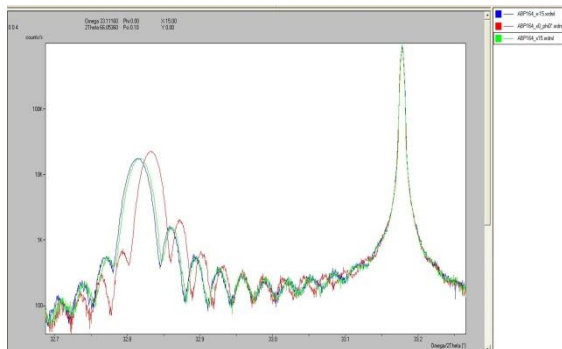


Fig. 2. X-ray diffraction scans for GaMnAs layer, in the center (red line) and to about 15 mm in the left and in the right of the center (blue and green lines).

The high resolution X-ray rocking curves around (004) reflection are shown in Fig. 2 for the annealed GaMnAs sample. The sharp peak 33.18° corresponds to the diffraction by the GaAs substrate. The GaMnAs diffraction is located in the lower angle side, compared to the GaAs peak, a_{per} is therefore larger than a_{sub} ; the GaMnAs sample is under compressive strain on GaAs, in this sample, $\Delta a/a_{\text{sub}} = 9400 \text{ ppm}$ is high indicating that the rather large amount of Mn atoms have been inserted in the matrix (close to 11.3 %). We further noticed that the lattice mismatch is more significant close to the edges of plate than in the center. We found 9900 ppm and 9820 ppm respectively to about 15 mm in the left and in the right of the center.

This result would therefore suggest that annealing favors a rather large reduction of the interstitial concentration in the following sample.

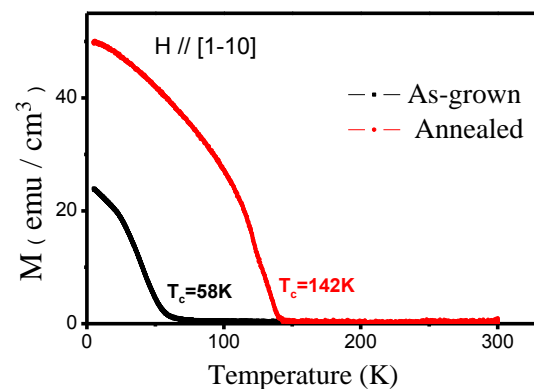


Fig.3. Magnetization curves for both as grown and annealed GaMnAs layer as a function of temperature. A static magnetic field of 500 Oe is applied for the as-grown layer and 100 Oe for the annealed one

The temperature dependence of the magnetization M (T) is shown in Fig.3. We applied a magnetic field in plane for the two layers ($H=500$ Oe for as-grown, and 100 Oe for annealed). The Curie temperature of As-grown 200 nm thick layers is typical for ferromagnetic GaMnAs samples [19], with a T_c about 58 K. Through the annealing technique, we note that T_c increases up to 142 K (Fig. 3). Indeed, this increase may therefore be related to the removal of Mn ions in GaMnAs that occupy interstitial sites (Mn_I). Since Mn_I acts as a donor and hence compensates the holes, optimal annealing increases the hole density and correspondingly enhances T_c . So we can improve the magnetic properties of this sample by reducing the amount of compensating defects [20].

In addition to the effect of annealing, an important clue to limitations on T_c in GaMnAs comes from the thickness of samples [20].

The effect of film thickness on the distribution of Mn atoms at various lattices sites in (Ga, Mn)As thin films have been investigated [21]. The authors quoted find that for film thickness less than 60 nm the growth surface acts as a sink facilitating the out diffusion of Mn interstitials Mn_I , and thus reducing its concentration in the film. The out diffused Mn_I accumulate on the surface layer and do not participate in the ferromagnetism of the film. For thin films less than 15 nm thick, no Mn_I can be detected. Because of the absence of compensating Mn_I defects, higher T_c can be achieved for such extremely thin $Ga_{1-x}Mn_xAs$ layers.

In a previous work, T_c 's exceeding the 110 K limit have been reported in thin $Ga_{1-x}Mn_xAs$ films (<50 nm thick) after

low temperature annealing [20,22]. Ku *et al.* [20] noted that, while they achieved a maximum T_c of 150 K for 20 nm $Ga_{1-x}Mn_xAs$ films, they did not succeed in achieving $T_c >110$ K (after annealing) for samples thicker than 50 nm. In very recent study on 150 nm GaMnAs sample with 10 % Mn doped, T. de Boer *et al.*[23] obtain a T_c about 135 K. In general, the lower T_c in thick sample was attributed for two reasons. Firstly, to the presence of Mn_I which electrically compensate the Mn_{Ga} acceptors and cancel the Mn_{Ga} Spins [24,25]. Secondly, the out diffusion of Mn_I to the surface was limited in thicker samples [21]. Here, we found a value after annealing of the Curie temperature up to 142 K, which is remarkable in GaMnAs in spite of a 200 nm thickness. This result shows the good quality of the sample used in this work and excludes that the out diffused of Mn_I accumulate on the surface layer is limited in the thick sample under investigation.

It is clear in previous work that the effect of the thickness is less pronounced in GaMnAs layer with Mn-doped above 10 %. Indeed Ben Hamida *et al.* [26] highlighted a reduction of this effect by the comparison between 25 nm and 100 nm GaMnAs films. We can notice that the high value of T_c in this layer is remarkable since it is comparable to those authors reference samples with small t . Chiba *et al.* [27] obtain T_c about 140 K for 4 nm GaMnAs layer with an annealing temperature $T_a=180^\circ$ C. Also Cubukcu *et al.* [28] reported a T_c about 130 K for an annealed 50 nm GaMnAs film with 10 percent of Mn. With 200 nm GaMnAs thick layer, we found a similar result to samples with small t and thus prove that the Mn out diffusion is yet not significantly thickness-limited.

4. Conclusion:

To summarize, the magnetic properties of thick $\text{Ga}_{0.9}\text{Mn}_{0.1}\text{As}$ film are reported using the VSM technique. We have found that the post-growth annealing results in a dramatic enhancement of both M_s and T_c comparable with GaMnAs layers with small thicknesses. This result highlights the good quality of the sample and shows that the out diffusion mechanism is not limited to a thickness up to 200 nm under investigation contrary to the previous conclusion [21].

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