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In-plane uniaxial anisotropy rotations in (Ga, Mn)As thin films

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Using SQUID magnetometry have found that in (Ga, Mn)As films the in-plane uniaxial magnetic easy axis is consistently associated with particular crystallographic directions. It can be rotated from the $[\bar{1}10]$ direction to the [110] direction by low temperature annealing and we show that this change is hole density-related. We demonstrate that the magnitude of uniaxial anisotropy as well its dependence on hole-concentration and temperature can be explained in terms of the p-d Zener model of ferromagnetism assuming a small trigonal-like distortion.

Key words*: GaMnAs; magnetic anisotropy; ferromagnetic semiconductor*

1. Introduction

Diluted magnetic semiconductors (DMS) in general and (Ga, Mn)As in particular are semiconducting materials for which a few percent of a magnetic element is added to a magnetically inert host. The discovery of carrier-mediated ferromagnetism in (III, Mn)V and (II, Mn)VI DMS grown by molecular beam epitaxy makes it possible to examine the interplay between the physical properties of semiconductor quantum structures and ferromagnetic materials [1] and opens the door for their application in spintronics, where electron spin is used to carry information [2]. It has been known since the early works in the field that (Ga, Mn)As films show strong magnetic anisotropy, which is largely controlled by epitaxial strain. The magnetic easy axis orients out-of-plane and in-plane under tensile and compressive biaxial strains, respectively

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[3-5], which is well understood within the framework of the p-d Zener model of hole-mediated ferromagnetism [6–8]. More recent works have shown that these systems also exhibit a strong in-plane uniaxial magnetic anisotropy [5, 8–10], pointing to the existence of a symmetry breaking mechanism, whose microscopic origin has not been identified yet. Since magnetic anisotropy will have a marked influence on spin injection and magnetotunnelling devices, it is important to develop a greater understanding of this property, and methods for its control. In this paper, we summarise the results of a study of in-plane magnetic anisotropy in a series of 50 nm thick (Ga, Mn)As thin films with Mn concentration x ranging from 1.7 to 9%.

2. Experimental

The crystallographic orientation of the wafer was determined from RHEED measurements during growth. For magnetometry studies, the material was typically cleft into 4×5 mm^2 rectangles, whose precise crystallographic orientation was reconfirmed by Laue back-reflection X-ray diffraction. Hole concentration was altered by low-temperature annealing. This procedure promotes the out-diffusion of compensating Mn interstitials from the layers [11], thus leading to an increased hole concentration and a higher Curie temperature [12]. Magnetic anisotropy was assessed in a custom-built low-field SQUID magnetometer. Detailed information about magnetic anisotropy was obtained from $M(H)$ curves recorded for various crystallographic orientations [13] and from the temperature dependence of the remnant magnetization M_{REM} [14].

3. Results and discussion

Magnetization loops for two representative samples with Mn concentrations of 2.2% and 5.6% are shown in Fig. 1a–d. It is well established that for typical hole densities (Ga, Mn)As/GaAs exhibits an in-plane magnetic anisotropy, determined by the superposition of two components, a biaxial, cubic-like anisotropy with [100] being the easy axes, and an uniaxial anisotropy with [110] being the easy axes [5, 8–10]. The former is a direct consequence of the spin anisotropy of the hole liquid, originating from strong spin-orbit coupling in the valence band. This coupling transfers all the complexities of valence band physics into the Mn ion subsystem. The second in-plane anisotropy component, the uniaxial term, is not expected from the above model, since its presence is precluded by general symmetry considerations in the biaxially strained zinc blende structure of (Ga, Mn)As. It has been suggested that these uniaxial properties may originate from symmetry lowering ($D_{2d} \Rightarrow C_{2v}$) due to either (i) a lack of top-bottom symmetry in (Ga, Mn)As epilayers [8] or (ii) anisotropic GaAs surface reconstruction during growth [11]. Importantly, it is usually found that the cubic term is dominant only at low temperatures. At elevated temperatures, the uniaxial term becomes larger than the cubic one. In such a case, the magnetic anisotropy of the layers is solely determined by the uniaxial term. Therefore, we focus on this high temperature regime which allows an unambiguous determination of the easy magnetic axis. We note (see Fig. 1a, b) that for both as-grown samples a clear uniaxial behaviour with $[\bar{1}10]$ being the easy axis is indeed observed.

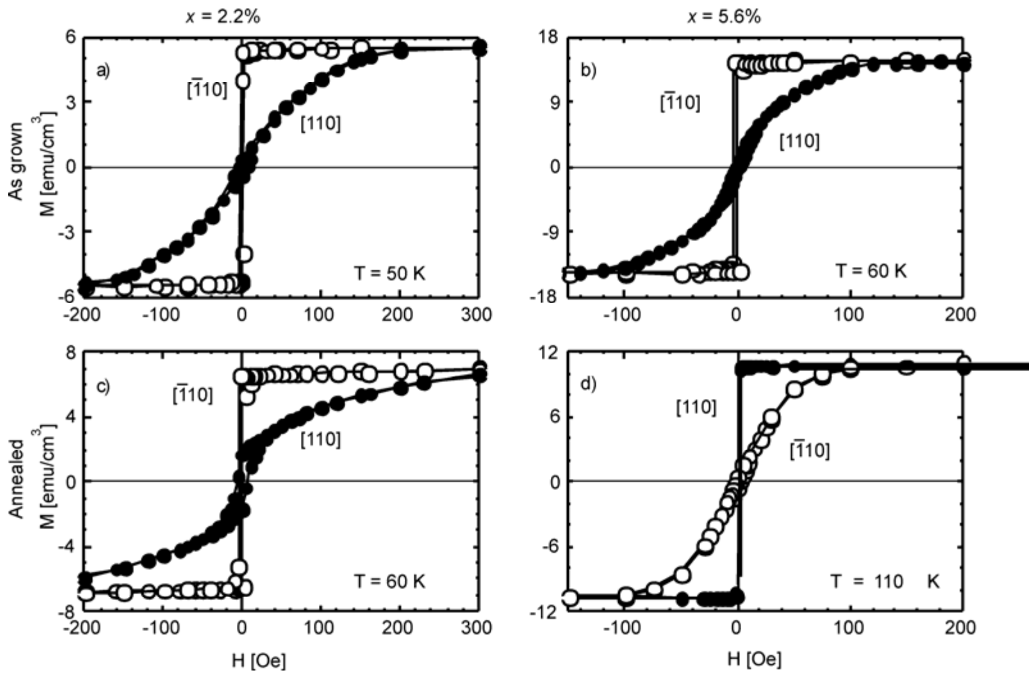

Fig. 1. Magnetization curves at temperatures close to T_C measured for [110] and $[\bar{1}10]$ orientations before a), b) and after annealing c), d) for $Ga_{0.978}Mn_{0.022}As$ and $Ga_{0.944}Mn_{0.056}As$. Note the switch of the magnetic easy axis from $[\bar{1}10]$ to [110] upon annealing for the latter sample

A similar behaviour is observed in all studied as-grown samples. At low Mn concentrations, annealing results in a relatively small increase in T_C and has no qualitative effect on magnetic anisotropy, as indicated in Fig. 1a, c for $x = 2.2\%$. A very different behaviour is observed at higher concentrations. As shown in Fig. 1b, d for $x = 5.6\%$, after annealing the easy axis rotates to the [110] direction. Before annealing, the $M(H)$ curves are square along the $[\bar{1}10]$ direction and elongated along the [110] direction, and this situation is reversed after annealing. Interestingly, some annealed samples also exhibit a temperature-induced reorientation of the easy axis [14]. In this case, the uniaxial easy axis points in the $[\bar{1}10]$ direction at low temperatures, but assumes the [110] direction at higher temperatures.

Since the annealing-induced increase of hole density is larger for the 5.6% sample, we ascribe the rotation of the easy magnetic axis to the increasing hole concentration. This modifies the relative occupancies of the valence sub-bands of the GaAs host, which (at least in the case of cubic anisotropy) makes competing contributions to the magnetic anisotropy [7]. Figure 2a shows the measured hole concentration in fuction of Mn concentration for the series of studied samples. By a close inspection of the figure, we can assign a threshold value of p of approximately 6×10^{20} cm^{-3}, above which the easy axis orients along [110]. An independent study showed that the uniaxial anisotropy field is thickness-independent in the range 0.2 to 6.8 μm [15]. Both findings strongly indicate that the in-plane uniaxial anisotropy depends on the bulk film parameters. Thus, this anisotropy can be caused by a symmetry lowering mechanism existing inside the film.

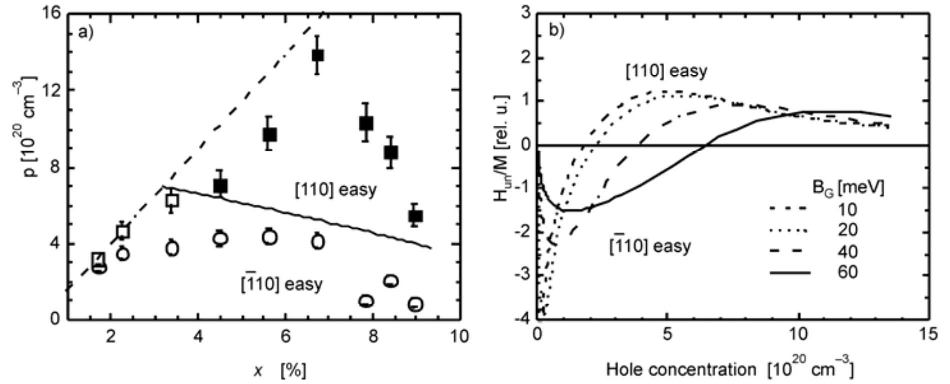

Fig. 2. The uniaxial easy axis fan chart for (Ga, Mn)As. Circles – as grown, squares – after annealing. Open symbols mark samples with the uniaxial easy axis oriented along the $[\bar{1}10]$ direction, full symbols denote samples exhibiting the easy axis along [110]. The dashed line marks the compensation-free p-type Mn doping level in (Ga, Mn)As. The solid line separates two regions of hole densities where, independently of being annealed or not, at elevated temperatures the layers consistently show the same crystallographic alignment of the uniaxial easy axis (a). In-plane uniaxial anisotropy field (normalized to sample saturation magnetization) versus hole density computed for various valence-band spin-splitting in (Ga, Mn)As assuming a non-zero trigonal distortion (b)

In order to find out the magnitude of the symmetry lowering perturbation that would explain our findings, we incorporate a trigonal distortion in the p-d Zener theory of ferromagnetism in tetrahedrally coordinated semiconductors [6, 7] described by the deformation tensor component $\varepsilon_{xy} \neq 0$. The computed anisotropy field corresponding to the in-plane uniaxial anisotropy H_{un} is shown in Fig. 2b as a function of hole concentration and parameterised by the valence-band spin-splitting parameter $B_G = A_F\beta M(T)/6g\mu_B$, where $g = 2.0$, $A_F = 1.2$ is the Fermi liquid parameter, and $\beta = -0.054$ eV·nm^3 is the p-d exchange integral. In the relevant region of hole concentrations and for ε_{xy} =0.05%, H_{un} attains experimentally observed values ≤ 0.1 T. Moreover, for a given value of x, our calculations confirm the possibility of the easy axis rotating from $[\bar{1}10]$ to [110] with increasing hole density. Additionally, since M and B_G are decreasing functions of T, the experimentally reported uniaxial easy axis reorientation transition ($[\bar{1}10] \Rightarrow [110]$) with increasing T is also well explained. Therefore, the $\varepsilon_{xy} \neq 0$ model qualitatively reproduces the observed change in the easy axis direction as a function of both hole concentration and temperature. The distortion required by the model may be associated with magnetostriction, or may result from a non-isotropic Mn distribution, caused for instance by the presence of surface dimers oriented along the $[\bar{1}10]$ direction during epitaxy. Since it has not yet been seen in other experiments, we conclude that the decisive microscopic mechanism that breaks the D_{2d} symmetry of (Ga, Mn)As epitaxial films is still to be found.

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