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

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We show, by SQUID magnetometry, that in (Ga,Mn)As films the in-plane uniaxial magnetic easy axis is consistently associated with particular crystallographic directions and that it can be rotated from the [110] direction to the [110] direction by low temperature annealing. We show that this behavior is hole-density-dependent and does not originate from surface anisotropy. The presence of uniaxial anisotropy as well its dependence on the hole-concentration and temperature can be explained in terms of the p-d Zener model of the ferromagnetism assuming a small trigonal distortion.

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Carrier-mediated ferromagnetism in the magnetic semiconductor (Ga,Mn)As has attracted considerable interest due to its potential application in spintronics, where the electron spin is used to carry information. It has been known since early works that (Ga,Mn)As films show rather 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. This is well understood within the framework of the p-d Zener model of the hole-mediated ferromagnetism. More recent works have shown, that these systems also exhibit a strong in-plane uniaxial magnetic anisotropy. This shows the existence of a symmetry breaking mechanism, whose microscopic origin has not yet been identified. Since the magnetic anisotropy will have a marked influence on spin injection and magnetotunneling devices, it is important to develop a greater understanding of this property, and the methods for its control.

Here we study the in-plane magnetic anisotropy in a series of (Ga,Mn)As thin films, and show that the easy direction is either [110], [100], or [110], depending on the carrier density and also on the temperature. We demonstrate, in particular, that the orientation of the uniaxial axis changes by 90° after annealing, if the hole concentration is increased above \( \sim 6 \times 10^{20} \text{ cm}^{-3} \). This finding suggests the in-plane magnetization direction can be controlled by changing the hole concentration, e.g. by gating or illumination. Furthermore, studies of magnetic response after subsequent etching steps shows that the uniaxial anisotropy is not a surface effect. By extending the previous theory to the case of arbitrary deformation, we find that the observed in-plane uniaxial anisotropy can be accounted for by a trigonal distortion \( \varepsilon_{xy} \approx 0.05\% \).

A wide range of 50 nm thick Ga\(_{1-x}\)Mn\(_x\)As thin films with \( 1.7 \leq x \leq 9\% \), and of different thickness (\( 10 \leq d \leq 100 \text{ nm} \)) for fixed Mn concentration \( x = 6.7\% \) were grown on GaAs(001) substrates by low temperature (180–300°C) molecular beam epitaxy using As\(_2\). The Mn concentrations are determined from the Mn/Ga flux ratio, calibrated by secondary ion mass spectroscopy (SIMS) on 1 \( \mu \text{m} \) thick samples grown under otherwise identical conditions. The crystallographic orientation of the wafer is determined from RHEED measurements during growth, with measurements along the [110] direction giving the 2\( \times \) pattern. For the magnetometry studies the material is cleft into, typically, 4 \( \times \) 5 mm\(^2\) rectangles, whose precise crystallographic orientation is reconfirmed by Laue back-reflection x-ray diffraction, which can unambiguously distinguish between diagonal [110] and [110] directions in GaAs. For electrical investigations, Hall bar structures are defined lithographically.

The hole concentration \( p \) is obtained from magnetoresistance and Hall measurements in magnetic fields up to 16 T and temperatures down to 0.3 K, using the routine described in detail previously. All samples show metallic conductivity, so that the hole concentrations can be obtained reasonably accurately by this method. The magnetic anisotropy is assessed using a custom built low-field SQUID magnetometer. Detailed information about magnetic anisotropy is obtained both from \( m(H) \) curves recorded for various crystallographic orientations and from the temperature dependence of the remnant magnetization \( M_{\text{REM}} \). For the latter the sample is cooled down through \( T_c \) in an external magnetic field of 1000 Oe, which is at least few times above any coercive field in the studied material. Then the field is removed at \( T = 5 \text{ K} \) and the \( M_{\text{REM}} \) component along the field-cooled direction is measured as a function of increasing temperature. The extremely low value of the external field trapped in our superconducting coil (typical trapped field stays below 0.1 Oe after a trip to 1000 Oe) allows the sam-
ple magnetization to rotate exactly to the nearest easy direction set only by the torques exerted on $M$ by internal anisotropy field(s). In general, this procedure, if repeated for the main crystallographic orientations, allows unambiguous determination of the orientation of $M$ across the whole temperature range up to $T_C$. Following the electrical and magnetic measurements, the samples are annealed at 190°C in air and then re-measured. Low temperature annealing is a well-established procedure for promoting outdiffusion of compensating Mn interstitials from the layers thus resulting in an increased hole concentration and Curie temperature.

Remanent magnetization $M_{\text{REM}}(T)$, for two representative samples with Mn concentration 2.2% and 5.6% respectively, are shown in Figs. 1 and 2. It is well established that for typical hole densities (Ga,Mn)As/GaAs exhibits an in-plane magnetic anisotropy which is determined by the superposition of two components: a biaxial, cubic-like anisotropy with [100] the easy axes, plus a uniaxial anisotropy with [110] the easy axes. The former is a direct consequence of the spin anisotropy of the hole liquid, originating from a strong spin-orbit coupling in the valence band. This coupling transfers all the complexities of the valence band physics into the Mn ions subsystem. As a result the strength of the cubic anisotropy strongly depends on epitaxial stress, hole concentration and temperature. 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 the symmetry lowering ($D_{2d} \rightarrow C_{2v}$) due to the lack of top-bottom symmetry in (Ga,Mn)As epilayers or as a consequence of the anisotropic GaAs surface reconstruction during growth.

It is usually found that the cubic term is dominant at low temperatures. However, on increasing temperature the strength of the uniaxial term decreases much more slowly than that of the cubic one (the uniaxial anisotropy field, $H_U$, decreases as $M$ while the cubic one, $H_C$, falls as $M^3$), and at elevated temperatures the uniaxial term becomes firstly comparable with, and soon after, stronger than the cubic contribution. In the latter case the magnetic anisotropy of the layers is solely determined by the uniaxial term. Note that the temperature corresponding to the crossover from cubic to uniaxial behaviour will be determined by the hole density and the precise form of $M(T)$. This crossover is not necessarily close to $T_C/2$. In the present study we focus on this high temperature regime, in which $M_{\text{REM}}$ is firmly locked into the easy magnetic direction, which allows an unambiguous determination of the easy magnetic axis.

The data of Figs. 1(a) and 2(a) clearly show that the magnetization is locked into the [110] direction for a wide temperature range below $T_C$ for the as-grown samples: along this direction $M_{\text{REM}}$ is the largest, and Brillouin-like; $M_{\text{REM}}$ along [110] is (very) small, $M_{\text{REM}}$ along [100] is identical with [010] and both are reduced by approximately $\cos(45^\circ)$. This shows that the uniaxial anisotropy is dominant at these temperatures, as discussed above. A similar behavior is observed in all as-grown samples studied here.

At low Mn concentrations, annealing results in a relatively small increase in $T_C$ and has no qualitative effect on the magnetic anisotropy, as shown in Fig. 1(b) for $x = 2.2\%$. A very different behavior is observed at higher concentrations. As shown in Fig. 2 for $x = 5.6\%$, after annealing the easy axis is rotated by $90^\circ$ to the
The face of the layers in $H_{[110]}$ origin of the observed reorientation by etching the surface magnetic anisotropy. However, we rule this out as the Curie temperatures.

In this case, the easy axis points in the $[\bar{1}10]$ direction, and this situation is reversed after annealing. Interestingly, we also observe the temperature-induced reorientation of the easy axis as shown in Fig. 3.

We therefore ascribe the rotation of the easy magnetic axis to the increase of the hole concentration on annealing. This will modify the relative occupancies of the valence sub-bands of the GaAs host, which (at least for the case of cubic anisotropy) make competing contributions to the magnetic anisotropy. Figure 5 plots the measured hole concentration versus the Mn concentration for the series of 50 nm thick samples. Open symbols mark samples where the easy axis is oriented along the $[110]$ direction, while for closed symbols the easy axis is along $[\bar{1}10]$. It can be seen that the easy axis rotates by $90^\circ$ on annealing for samples with $x \gtrsim 4\%$. By inspection of Fig. 5, we can assign a threshold value of $p$ of approximately $6 \times 10^{20}$ cm$^{-3}$, above which the easy axis orients along $[110]$. This finding, together with the etching experiment discussed above demonstrate that the in-plane uniaxial anisotropy depends upon the bulk film parameters. A recent study showing that the uniaxial anisotropy is thickness-independent in the range 0.2 $\mu$m to 6.8 $\mu$m gives further support to this argument. Thus, this anisotropy is caused by a symmetry lowering mechanism existing inside the film.

In order to find out the magnitude of the symmetry lowering perturbation that would explain our findings we incorporate in the p-d Zener theory of ferromagnetism in tetrahedrally coordinated semiconductors a trigonal...
a switch in the easy axis from [110] to [110] will occur on increasing hole density. As $M$, and so $B_G$, are decreasing functions of $T$, a uniaxial easy axis reorientation transition $[110] \Rightarrow [110]$ may occur also on increasing $T$, see Fig. 6. Therefore the model qualitatively reproduces the observed change in the easy axis direction as a function of both hole concentration and temperature.

In summary, we have shown that the easy magnetization axis of (Ga,Mn)As films can rotate from the [110] direction to the [110] direction on annealing. We demonstrate that the orientation of the in-plane uniaxial anisotropy in (Ga,Mn)As is dependent on the hole concentration. Since the hole concentration in III-V magnetic semiconductors can be modified by gating or illumination, this suggests the possibility of a 90° rotation of the magnetization without application of an external magnetic field, which could have important applications in spintronics. Our results show that this behavior does not originate from surface anisotropy. We demonstrate that the magnitude of uniaxial anisotropy as well its dependence on the hole-concentration and temperature can be explained in terms of the p-d Zener model of the ferromagnetism assuming a small trigonal–like distortion. Such a distortion may be associated with magnetostriiction, or may result from a non-isotropic Mn distribution, caused for instance by the presence of surface dimers oriented along [110] direction during the epitaxy. However, the dominating microscopic mechanism that breaks $D_{2d}$ symmetry of (Ga,Mn)As epitaxial films is to be elucidated.

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