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Magnetic Properties of (Ga,Mn)As

M. SAWICKI*

Institute of Physics, Polish Academy of Sciences al. Lotników 32/46, 02-668 Warsaw, Poland

A review is given of experimental findings and theoretical understanding of micromagnetic properties of zinc-blende ferromagnetic semiconductors, with (Ga,Mn)As taken as a sole example. It is emphasised that the Zener p-d model explains quantitatively the effect of strain on the easy axis direction as well as it predicts correctly the presence of the reorientation transition, observed as a function of hole concentration and temperature. Possible suggestions put forward to explain the existence of in-plane uniaxial magnetocrystalline anisotropy are then quoted.

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1. Introduction

The discovery of carrier-mediated ferromagnetism in (III,Mn)V and (II,Mn)VI dilute magnetic semiconductors (DMS) grown by molecular beam epitaxy makes it possible to examine the interplay between physical properties of semiconductor quantum structures and ferromagnetic materials [1, 2]. At the same time, complementary resources of these systems open doors for novel functionalities and devices. A considerable effort in this field is focused on identifying methods for mutual manipulations of semiconductor and magnetic properties as well as on developing DMS, in which the ferromagnetism can persist above room temperature. In this context (Ga,Mn)As serves as a valuable test ground for DMS properties, due to the relatively high $T_{\rm C}$ and its compatibility with the well--characterised GaAs system. The Mn dopant in this III-V host matrix is expected to substitute for the Ga site, and fulfil two roles: to supply a local spin 5/2 magnetic moment, and to act as an acceptor, providing itinerant holes which mediate the ferromagnetic order. The theoretical understanding of this phenomenon [3] is built on Zener's model of ferromagnetism, the Ginzburg–Landau approach to the phase transitions, and the Kohn–Luttinger kp theory of semiconductors. Within

^{*}e-mail: mikes@ifpan.edu.pl

this model and its variants, the magnitude of the Curie temperature $T_{\rm C}$ in Mndoped GaAs, InAs, GaSb, InSb [4–6] as well as in *p*-CdTe, *p*-ZnTe, and Ge [7] is understood assuming that the long-range ferromagnetic interactions between the localised spins are mediated by delocalised holes in the weakly perturbed valence band [8]. The assumption that the relevant carriers reside in the *p*-like valence band makes it possible to describe various magnetooptical [4, 9] and magnetotransport properties of (Ga,Mn)As, including the anomalous Hall effect and anisotropic magnetoresistance [9, 10] as well the negative magnetoresistance caused by the orbital weak-localisation effect [11]. From this point of view, (Ga,Mn)As and related compounds emerge as the best understood ferromagnets, providing a basis for the development of novel methods enabling magnetisation manipulation and switching [12].

Here, a brief review of micromagnetic properties of (Ga,Mn)As is given. Interestingly, despite much lower spin and carrier concentrations compared to ferromagnetic metals, (III,Mn)V exhibit excellent micromagnetic characteristics, including well-defined magnetic anisotropy and large ferromagnetic domains separated by usually straight-line domain walls. It turns out that the above-mentioned p-d Zener model explains the influence of strain on magnetic anisotropy as well as describes the magnitudes of the anisotropy field and domain width. Importantly, the experimentally observed reorientation transition as a function of the temperature and hole concentration is readily accounted for. At the same time an additional weak in-plane magnetic anisotropy that has been detected in these systems points to a symmetry breaking, whose origin has not yet been identified.

2. Curie temperature

Ferromagnetic ordering of the relatively widely-spaced Mn dopants in the semiconductor host arises from antiferromagnetic exchange interactions between Mn 3d magnetic moments and the delocalised charge carriers. The wide ranging experimental studies of (Ga,Mn)As of the past few years have revealed several curiosities which triggered intensive theoretical debate, namely: (i) the hole density p is often much smaller than the Mn density x [13]; (ii) the saturation magnetisation M_{sat} may be smaller than the expected 4–5 μ_{B} per Mn atom [14, 15]; (iii) the Curie temperature $T_{\rm C}$ saturates or even tails off as x is increased above around 5% [13, 15]. These peculiarities proved to be inherently related to growth of (Ga,Mn)As with the Mn and hole concentrations surpassing thermal equilibrium limits by use of low temperature epitaxy. This leads to a high density of defects with As antisites, As_{Ga} , and interstitial Mn, Mn_I , being the most numerous. The crucial role of the latter has been proven by demonstrating that (iv) p, $T_{\rm C}$, and $M_{\rm sat}$ can all be increased by annealing at temperatures comparable to [15, 16] or even lower than [17] the growth temperature. Since Mn_I is a double donor in (Ga,Mn)As, it compensates holes provided by substitutional Mn_{Ga} .

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Further, tight-binding [18] and density-functional [19] calculations indicate that Mn_{I} couples antiferromagnetically to neighbouring Mn_{Ga} , which combined with the former effect suppresses ferromagnetism even further. So at least two steps have been identified to minimise defect densities: careful control of growth [20] followed by the post growth annealing.

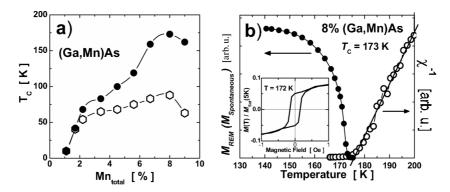


Fig. 1. (a) Ferromagnetic transition temperatures versus total Mn concentration for asgrown (open) and annealed (filled) (Ga,Mn)As thin films; (b) temperature-dependence of remnant magnetisation and inverse paramagnetic susceptibility for annealed 8% sample; the inset: hysteresis loop for the same sample at 172 K.

Figure 1 shows results of magnetic investigations performed on such a carefully prepared set of (Ga,Mn)As layers. In particular, the growth temperature is chosen to be the highest possible while maintaining 2D growth (Mn flux depending), with As flux kept low in order to minimise the concentration of compensating As_{Ga} [20]. The total Mn concentration is determined from the Mn/Ga flux ratio, which was calibrated by SIMS measurements on 1 μ m thick samples grown under otherwise the same conditions. Magnetisation studies are performed on a home made SQUID magnetometer dedicated to low magnetic field studies of minute signals expected for layered DMS structures. A special care is devoted to screen the sample from external fields, and to keep the parasite remnant fields generated by the magnet at the lowest possible level (usually just below 0.1 Oe). Figure 1a shows the ferromagnetic transition temperatures $T_{\rm C}$ in 25–50 nm thick (Ga,Mn)As layers as-grown and annealed at 190°C for several tens of hours. $T_{\rm C}$ is obtained from the temperature-dependence of the remnant magnetisation measured by SQUID, which agrees within 2 K accuracy with anomalous Hall measurements using Arrott plots. The annealing clearly has a pronounced effect on $T_{\rm C}$, especially at high Mn concentrations. This is a clear manifestation of the removal of compensating interstitial Mn from the bulk of the layers to the free surface, where they get passivated [17, 19, 21, 22]. The resistivity and hole density show similar trends [23].

The highest $T_{\rm C}$ so far is 173 K, a value obtained for an annealed 25 nm thick sample with nominal 8% of Mn [24]. To the authors knowledge this is the highest

value reported in (Ga,Mn)As single layers. The remnant magnetisation and inverse paramagnetic susceptibility versus temperature for this sample is shown in Fig. 1b. To further confirm this finding a clear ferromagnetic hysteresis at 172 K for this sample is shown in the inset.

3. Origin of magnetic anisotropy

The magnetic dipolar anisotropy, or shape anisotropy, is mediated by the dipolar interaction. Since it is long range, its contribution depends on the shape of the sample and in thin films the shape anisotropy often results in the in-plane alignment of the moments. For thin films, the shape anisotropy energy per unit volume is given by

$$E = \frac{1}{2}\mu_o M_{\rm s}^2 \cos^2\theta,\tag{1}$$

which leads to the anisotropy field $\mu_o H_A = \mu_o M$ of about 60 mT for $Ga_{0.95}Mn_{0.05}As$ (M_s is the saturation magnetisation and θ is the angle the magnetisation subtends to the plane normal). Already early studies of the ferromagnetic phase in (In,Mn)As [25] and (Ga,Mn)As [26] demonstrated the existence of magnetic anisotropy, whose character and magnitude implied a sizable contribution of a microscopic origin. Namely, it has been found by studies of the anomalous Hall effect [26, 27] and ferromagnetic resonance [28] that the direction of the easy axis is rather controlled by epitaxial strain in these systems. Generally, for layers under tensile biaxial strain (like (Ga,Mn)As on an (In,Ga)As buffer) perpendicular-to--plane magnetic easy axis has been observed. In contrast, the layers under compressive biaxial strain (as canonical (Ga,Mn)As on a GaAs substrate) have been found to develop in-plane magnetic easy axis. At first glance this sensitivity to strain appears surprising, as the Mn ions are in the orbital singlet state ${}^{6}A_{1}$ [29]. For such a case the orbital momentum L = 0, so that effects stemming from the spin-orbit coupling are expected to be rather weak and, indeed, electron paramagnetic resonance studies of Mn in GaAs have led to relevant spin Hamiltonian parameters by two orders of magnitude too small to explain the values of $\mu_o H_A$ [30]. However, the interaction between the localised spins is mediated by the holes that have a non-zero orbital momentum l = 1 [3]. An important aspect of the p-d Zener model is that it does take into account the anisotropy of the carrier-mediated exchange interaction associated with the spin-orbit coupling in the host material [3, 4, 31].

In order to visualise the origin and direction of the expected magnetic anisotropy it is instructive to examine the electronic structure of the top of the valence band in biaxial strained zinc-blende compounds. In this case the valence band splits and the energetic distance between the heavy-hole $j_z = \pm 3/2$ and light-hole $j_z = \pm 1/2$ subbands depends on strain, see Fig. 2. For the biaxial *compressive* strain the ground state subband assumes a heavy-hole character. Then, assuming for the sake of simplicity that only the ground state subband is

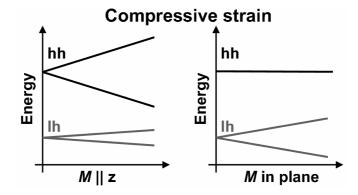


Fig. 2. Illustration of valence band splitting of tetrahedrally coordinated semiconductors for compressive strain and for two orientations of magnetisation M in respect to the sample plane.

occupied, the hole spins are oriented along the growth direction. Now, since the p-d exchange interaction has a scalar form, $H_{\rm pd} \sim s \cdot S$, the in-plane Mn spin magnetisation M will not affect the heavy-hole subband. This means that perpendicular magnetic anisotropy is expected, since only for such a magnetisation orientation the holes can lower their energy by the coupling to the Mn spins. In the opposite, *tensile* strain case, the in-plane component of the hole spin is greater than the perpendicular component, so a stronger exchange splitting will occur for the in-plane orientation of M. Hence, the in-plane anisotropy is expected if only the light-hole subband remains occupied. Exactly such a case we meet for quantum wells of modulation-doped p-type (Cd,Mn)Te grown under compressive and tensile strain [32]. As observed previously [33], for the compressive strain a ferromagnetic state related splitting of the luminescence line occurs only for the perpendicular orientation. However, when large enough tensile strain was built into this system, the in-plane direction of the easy axis has been observed.

By nature (III,Mn)V DMS systems are heavily populated with holes and such a simple model can serve only as a guide line. It must be noted therefore, that when the Fermi energy is comparable or even larger than the heavy-hole – light-hole splitting, the strong mixing takes place and we can only talk about either heavy- or light-hole like character of the holes.

4. Strain-induced anisotropy and reorientation transition

A detail theoretical analysis of anisotropy energies and anisotropy fields in films of (Ga,Mn)As requires elaborate numerical calculations and such ones have been carried out for a number of experimentally important cases within the p-d Zener model [4, 31]. In particular, the cubic anisotropy as well as uniaxial anisotropy under biaxial epitaxial strain have been examined as a function of the hole concentration and temperature. Computed reorientation lines of the mag-

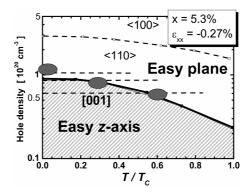


Fig. 3. Experimental (full points, taken from Fig. 4) and computed values of the ratio of the reorientation to Curie temperature for the perpendicular to in-plane magnetic anisotropy transition (thick line). Dashed lines mark expected temperatures for the reorientation of the easy axis between $\langle 100 \rangle$ and $\langle 110 \rangle$ in-plane directions.

netic easy axis for 5% (Ga,Mn)As/GaAs layer are presented Fig. 3. Both shape and magneto-crystalline anisotropies have been taken into account. Firstly, it should be noted that both in and out of plane magnetic anisotropy can be realised in this system. This is the direct consequence of the model sketched above. Accordingly, and quite generally, samples with at least moderate hole concentration will exhibit easy axis located in plane, since for (Ga,Mn)As/GaAs valence band assumes then the light-hole like character. The opposite happens for heavily compensated samples. For low $E_{\rm F}$ values the magnetisation is favored out of the plane. So, according to the model, and despite common expectations, a low hole concentration (Ga,Mn)As will show perpendicular magnetic anisotropy. Secondly, the shape of the reorientation line allows for a change of the direction of the magnetic easy axis not only as a function of the hole density (isothermal, vertical crossing) but also via horizontal crossing, that is as a function of the temperature (this comes into play by controlling the magnitude of spontaneous magnetisation, and hence the spin splitting). Thirdly, it is worth emphasising the relatively narrow range of hole densities for which the reorientation may take place. So, if the hole concentration for a given strain, is either too small or too large no reorientation transition is expected for any value of magnetisation (temperature) changes. On the other hand, for an appropriate combination of strain and the hole density, even a minute change of temperature (magnetisation) switches the easy axis between the two directions.

Figure 4 depicts the pertinent experimental results for 5.3% (Ga,Mn)As/GaAs family of samples originating from the same wafer with increasing hole density prepared by the low-temperature annealing [34]. The figure collects the in-plane (the left panel) and perpendicular (the right panel) components of spontaneous magnetisation measured as a function of temperature

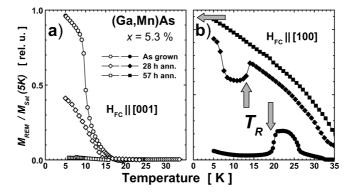


Fig. 4. Temperature dependence of the remanent magnetisation as measured in perpendicular [001] (a) and in-plane (100) (b) configurations for a Ga_{0.947}Mn_{0.053}As sample prior to annealing (circles) and after annealing (diamonds and squares). The sample is cooled down through $T_{\rm C}$ in the field $H_{\rm FC} = 0.1$ T, which is at least by a factor of ten higher than the coercive field $H_{\rm c}$. Then, the field is removed at 5 K, and the measurement of the magnetisation component M along the direction of $H_{\rm FC}$ commences on increasing temperature in the residual field $H_{\rm r} < 10 \ \mu$ T. Note that the development of the in-plane component of M is accompanied by an equivalent quench of the perpendicular one. Bulk arrows mark the reorientation temperature $T_{\rm R}$ when the cross-over to in-plane magnetic anisotropy takes place (after Sawicki et al. [34]).

for these samples. Starting with the sample having the lowest hole density, the as-grown one, the strong out of plane and negligible small in-plane components observed at low temperatures instruct us that indeed the layer exhibits perpendicular magnetic anisotropy. This however reverses at elevated temperatures: both components swap their relative intensities above the reorientation temperature, $T_{\rm R}$, defined as the temperature above which a sizable increase of the in-plane components is observed. Direct measurements of magnetic hysteresis loops below and above $T_{\rm R}$ for both experimental configurations further confirm the existence of the magnetic anisotropy reorientation transition [34–36]. Remarkably, the opposite behaviour occurs in tensile strained (In,Mn)As/GaAs, where the easy axis switches from in-plane to out of plane on warming [37], so further endorsing the model.

This set of samples allows us to track the influence of p on $T_{\rm R}$ too. A clean decrease in $T_{\rm R}$ is observed upon annealing with $T_{\rm R}$ being finally pushed below the experimental temperature range for the layer with the higher hole density. So, this sample exhibits only in-plane magnetic anisotropy. The experimentally established $T_{\rm R}$ values are compared with the theoretical calculations performed for this particular sample, see Fig. 3. In view that the theory is developed with no adjustable parameters the agreement between experimental and computed pand T corresponding to the reorientation transition is very good.

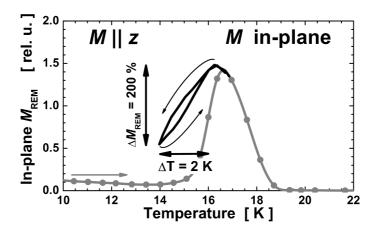


Fig. 5. Temperature cycling of the remnant in-plane magnetisation in 1% (Ga,Mn)As near the reorientation point (after Sawicki et al. [39]).

It is worth noting that although a change of the hole density by annealing as an irreversible process cannot be employed in any working device, p can be changed by applying an appropriate gate voltage [38], so a local control of the magnetic easy axis in ferromagnetic DMS seems feasible. Such an experiment has not been performed so far, but as Fig. 5 clearly demonstrates this possibility, when, instead of modulating p, a narrow range temperature cycling can produce reversible, almost on/off type, changes of the direction of the spontaneous magnetisation.

Finally, the experimentally found absolute values of biaxial strain induced uniaxial anisotropy fields [28, 33, 40, 41] remain also in very good agreement with the theoretical calculations if the large hole densities are assumed.

5. In-plane magnetic anisotropy

According to the discussion above, the easy axis assumes the in-plane orientation for typical carrier concentrations in (Ga,Mn)As/GaAs. In this case, according to the theoretical predictions presented in Fig. 5 as well in Fig. 9 of Ref. [4] and in Fig. 6 of Ref. [31] the fourfold magnetic symmetry with the easy axis is expected to switch between the $\langle 100 \rangle$ and $\langle 110 \rangle$ in-plane cubic directions as a function of p or T. This biaxial magnetic symmetry has indeed been observed at low temperatures, however with the easy axis assuming exclusively [100] in-plane orientations [28, 34, 40, 42–47]. To the authors best knowledge, no $\langle 100 \rangle \Leftrightarrow \langle 110 \rangle$ reorientation transition has been detected to date. It is possible that anisotropy of the hole magnetic moment, neglected in the theoretical calculations [4, 31], stabilizes the $\langle 100 \rangle$ orientation of the easy axis. However, whether such a model will explain simultaneously the reported recently $\langle 110 \rangle$ biaxial symmetry in (In,Mn)As/(In,Al)As films [48], remains to be shown. Nevertheless, the corresponding in-plane anisotropy field assumes the expected magnitude, of the order of 0.2 T at low temperatures, which is typically 2–3 times smaller than that corresponding to the strain-induced energy of magnetic anisotropy.

In addition to the cubic in-plane anisotropy, the accumulated data for both (Ga,Mn)As/GaAs [28, 34, 40, 42–47] and (In,Mn)As/(In,Al)As point to a non-equivalence of [110] and [–110] directions, which lead to the in-plane uniaxial magnetic anisotropy. As shown in Fig. 6 [34], remnant magnetisation M mea-

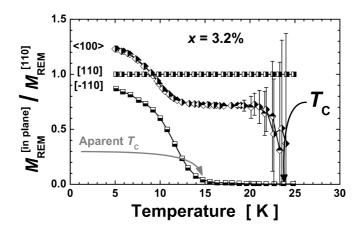


Fig. 6. Experimental evidence for the uniaxial anisotropy along [110] direction in $Ga_{0.97}Mn_{0.03}As$ film. The magnetic remanence is measured for four major in-plane directions and its magnitude is normalised by the data of the [110] case. Note that the sudden drop of M along [-110] at $T < T_C$ may wrongly indicate too low value of T_C , if only this orientation is probed (after Sawicki et al. [34]).

sured along [-110] direction vanishes completely above 15 K indicating that this is the hard direction in this film. We also note that when $M_{[-110]}$ vanishes, the $M_{[100]}/M_{[110]}$ ratio drops to $1/\sqrt{2}$, as expected for the easy axis along [110]. Since the cubic-like anisotropy energy is proportional to M^4 whereas the uniaxial one to M^2 , the latter though initially weaker is dominating at high temperatures, where M is small. Such a uniaxial anisotropy is not expected for D_{2d} symmetry of a T_d crystal under epitaxial strain. Furthermore, the magnitude of the corresponding anisotropy field appears to be independent of the film thickness, both for as large as 7 μ m [47] and as low as 25 nm [45] layers, which, in particular, rules out the effect of Mn oxide accumulated at the free surface [19, 49]. Initially, the present author with co-workers argued that a unidirectional character of the growth process and/or differences between (Ga,Mn)As/GaAs and (Ga,Mn)As/vacuum interfaces may lower symmetry to C_{2v} where the three principal directions are: [001], [110], and [-110]. They are not equivalent; in C_{2v} the $[110] \Leftrightarrow [-110]$ symmetry gets broken, while the $[100] \Leftrightarrow [010]$ one is maintained. The Argone–Notre Dame team recently has advocated for an effect connected with surface reconstruction induced preferential Mn incorporation occurring at every step of layer-by-layer

growth. However, none of the existing experimental findings obtained to date has provided any clue about this puzzling symmetry breaking in the film body.

6. Conclusions

In summary, experimental and theoretical findings discussed here demonstrate the rich characteristics of magnetic anisotropies in (Ga,Mn)As and related systems, which, in addition to epitaxial strain, vary with the hole and Mn concentrations as well as with the temperature. According to the theory [4, 31] these reflect spin anisotropy of the valence band subbands whose shape depends on strain, while the splitting and population on magnetisation and hole concentration.

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