Magnetic properties of (Ga,Mn)As

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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]. (Ga,Mn)As serves as a valuable test ground for ferro-DMS, due to the relatively high Curie temperature, T_C, and compatibility with the well-characterised GaAs system. The Mn dopants in this III-V host matrix are expected to substitute for the Ga site, and fulfill two roles: to supply a local spin 5/2 magnetic moments, and to act as an acceptor, providing itinerant holes which mediate the ferromagnetic order. The theoretical understanding of this phenomenon [2] is built on Zener's model of ferromagnetism, the Ginzburg-Landau approach to the phase transitions, and the Kohn-Luttinger $k \cdot p$ theory of semiconductors. Within this model, the magnitude of T_C in Mn-doped arsenides and antimonides, as well as in p-type tellurides and Ge is understood assuming that the long-range ferromagnetic interactions between the localized spins are mediated by delocalised holes in the weakly perturbed valence band [3]. The assumption that the relevant carriers reside in the p-like valence band makes it possible to describe various magnetooptical [4,5] and magnetotransport properties of (Ga,Mn)As, including the anomalous Hall effect and anisotropic magnetoresistance [5,6] as well the negative magnetoresistance caused by the orbital weak-localization effect [7]. 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 [8,9].

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 describing the magnitudes of the anisotropy field and domain width. Importantly, the experimentally observed various magnetic easy axis reorientation transitions as a function of the temperature and hole concentration are satisfactory accounted for. This applies also to a weak in-plane magnetic anisotropy that has been detected in these systems.

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 like lower hole density, p, than the Mn density x, or saturation magnetisation deficit for higher x. 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 structural defects, most importantly Mn interstitial [10]. It is not only that as a double charged center it removes two holes from the system, it also couples antiferromagnetically with neighbouring substitutional Mn [11,12]. It has been demonstrated however, that p, T_C, and M_{Sat} can all be increased by post growth annealing at temperatures comparable to [13] or lower than [14] the growth temperature. This is the manifestation of the removal of compensating Mn interstitials from the bulk of the layers to the free surface, where they get passivated [15,16].

Origin of magnetic anisotropy in ferromagnetic zinc-blende DMS

The magnetic dipolar anisotropy, or shape anisotropy, is mediated by 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 (in-plane magnetic anisotropy, IMA). One of the most intriguing findings concerning magnetic properties of (III,Mn)V ferro-DMS was the discovery of a perpendicular ferromagnetic order (perpendicular magnetic anisotropy, PMA) in p-(In,Mn)As [17]. Similar studies of the ferromagnetic phase in (Ga,Mn)As [18-20] also demonstrated the existence of PMA in some special cases. As all the layers are very thin (typically a fraction of μm thick) and of macroscopic lateral dimensions, the observation of PMA in these layers points to the existence of a strong, microscopic mechanism that counteracts the shape-imposed in-plane arrangement of the magnetisation.

Since the low temperature growth of (III,Mn)V precludes misfit dislocations formation, the resulting layers are pseudomorphic with respect to the GaAs substrate. This leads to large epitaxial (lattice-mismatch-driven) strain persisting well beyond the critical thickness. It is this epitaxial strain that sets the foundation for the magnetic anisotropy in ferro-DMS. It has been established that, generally, for compressive biaxial strain [as in canonical (Ga,Mn)As on a GaAs substrate] IMA develops. In contrast, for layers under tensile biaxial strain [like (Ga,Mn)As on an (In,Ga)As buffer] PMA is observed. The matter turns, however, to be more complicated. For example, the perpendicular orientation of spontaneous magnetisation can also be found in some (Ga,Mn)As/GaAs layers [21-23], that is when IMA is expected. So, it is clear that other factors (identified as hole concentration and temperature) play a role here, and that the resulting anisotropy is determined by a combination of all of them. Figure 1(a) gives a clear example of such a (Ga,Mn)As/GaAs layer that despite the strain-related expectations - exhibits PMA.

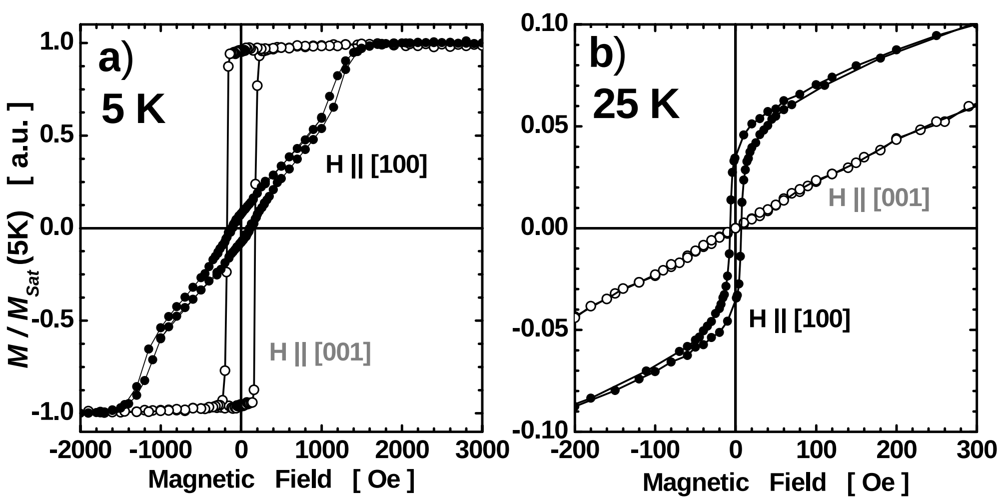

Fig. 1. Magnetic field loops for one $Ga_{0.977}Mn_{0.023}As$ film on GaAs substrate measured for two orientations with respect to the magnetizing field: full points - parallel, circles - perpendicular. At low temperature **(a)**, a perfect square hysteresis is obtained when the magnetic field is perpendicular to the film surface, while an elongated loop is seen when the in-plane orientation is probed. This clearly evidences that the easy axis is perpendicular. An opposite behaviour is observed at higher temperatures, **(b)**. The reversed character of the hysteresis loops indicates the flip of the easy axis direction between these two temperatures *(After Sawicki et al [21])*.

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 Figs. 6 and 9 of ref. [4] and in Fig. 6 of ref. [24] the fourfold magnetic symmetry is expected with the easy axis assuming either ⟨100⟩ and ⟨110⟩ in-plane cubic directions depending on *p* or *T*. This biaxial magnetic symmetry indeed is observed at low temperatures, however with the easy axis oriented exclusively along [100] in-plane orientation.

In addition to the cubic in-plane anisotropy, the accumulated data for both (Ga,Mn)As/GaAs [20,23,25-27] and (In,Mn)As/(In,Al)As [28] point to a non-equivalence of [110] and [-110] directions, which leads to the in-plane uniaxial magnetic anisotropy. Such a uniaxial anisotropy is not expected for D_{2d} symmetry of a T_d crystal under epitaxial strain. This shows the existence of a symmetry breaking mechanism, whose microscopic origin still needs to be identified. The accumulated data so far indicate that the magnitude of the corresponding anisotropy field appears to be independent of the film thickness, both for as large as 7 μm [27] and as low as 25 nm layers [29]. Also, a surface etching experiment ruled out an effect of Mn oxide accumulated at the free surface [16,30], and all together point strongly that the mechanism responsible for the uniaxial anisotropy may be a bulk property of the layered material.

It has been found experimentally, that (Ga,Mn)As layers respond to the external magnetic field as a single magnetic domain, encompassing the whole, macroscopic in size sample. Such a character of the IMA (Ga,Mn)As has been confirmed by direct magneto-optical domain mapping [26], and by a compliance to Stoner-Wohlfarth model of the coherent rotation in the presence of an external magnetic field [9,20,23,25-27,30,31]. In particular it has been found that the total energy of (Ga,Mn)As with in-plane magnetisation can be remarkably well described taking only the lowest order contributions of biaxial (in-plane cubic) and uniaxial terms. 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 [31]. Phenomenologically, it can be said that this property originates from the very long range nature of the Zener mechanism. On the microscopic level it has been shown [32] that the enhancement of the spatial uniformity of the spin ordering stems from p-like character of the valence band wave functions - a characteristic feature of zinc-blende (ferro-) DMS. When calculated within the framework of the model, the magnetic stiffness largely exceeds that expected for a simple spin degenerate band. Remarkably, these calculations correctly reproduce the stripe domains width of 1.5 μm, observed in demagnetised PMA (Ga,Mn)As [33].

Reorientations of magnetic easy axis

The scalar form of the exchange interaction in strained material dictates that Mn moments, or more accurately their collective macroscopic magnetisation, adjusts its orientation with respect to strain to minimize the total energy of the carriers required to support ferromagnetic ordering of the Mn ions. In particular, when only biaxial (in-plane) epitaxial strain is present the valence band splits and the energetic distance between the heavy-hole j_z = 3/2 and light-hole j_z = 1/2 subbands developes. For the biaxial compressive strain the ground state subband assumes a heavy-hole character. If then, *only this* state is occupied, the hole spins are oriented along the growth direction, the magnetisation is expected to follow, the PMA is established. However, depending on the Fermi level position within the valence band and/or the value of the exchange splitting (that is depending on magnetisation and thus also on temperature) different orientations of magnetisation can be required to drive the system to its energy minimum. Therefore, by changing hole concentration or temperature, the corresponding changes of the overall orbital momentum of the hole liquid may force a spontaneous reorientation of magnetisation, preset at the first place by the direction of the epitaxial strain alone.

It is easy to realise how the character of magnetic anisotropy depends on the temperature if we recall that the spin-splitting is proportional to Mn magnetisation *M(T)* that varies according to the Brillouin-like function. It immediately follows the temperature induced changes in the hh/lh populations may require corresponding magnetisation reorientations in order to sustain the ferromagnetic state. Accordingly, in compressively strained structures PMA occurs at both low temperatures and hole concentrations, while otherwise IMA will be realized. This implies that there exists a class of samples for which the material parameters are such that within the experimental temperature range the reorientation of the easy axis from easy z-axis to easy plane occurs on increasing temperature, see Fig. 1.

Similarly to the case of epitaxial strain induced anisotropy, the recent investigations [29] have shown that also uniaxial in-plane anisotropy shows pronounced sensitivity to p, x, and T. In particular, it has been established that the uniaxial easy axis of (Ga,Mn)As films is associated with particular crystallographic axes and that it can rotate 90^o from the [-110] to the [110] direction on annealing. Moreover, for a specific combination of p and x the uniaxial easy axis is found to be temperature dependent too.

Conclusions

In summary, the material presented here demonstrates how rich characteristics of magnetic properties are obtained if the magnetic subsystem is almost exclusively controlled by spin anisotropy of the valence band subbands. Remarkably, the model calculations [4,24], taking into account the influence of strain on the valence subbands shape, and the influence of hole density and temperature on the splitting and population, are found not only to correctly describe the emerging general picture, but also to quantitatively reproduce the relevant experimental findings. From one point of view, such a great sensitivity to minute details of the growth procedure or external parameters may spell some problems for utilisation of the hole mediated ferromagnets, on the other hand it opens a sea of new functionalities and makes ferro-DMS an extremely attractive subject of investigations.

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