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2004 J. Phys.: Condens. Matter 16 S5471

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Magnetic anisotropy and domain structure in carrier-controlled ferromagnetic semiconductors

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Received 1 June 2004

Published 19 November 2004

Online at stacks.iop.org/JPhysCM/16/S5471

doi:10.1088/0953-8984/16/48/001

Abstract

A review is given of the experimental situation and theoretical modelling of micromagnetic properties of zinc-blende ferromagnetic semiconductors, such as (Ga, Mn)As and p-(Cd, Mn)Te. It is emphasized that the Zener p–d model explains quantitatively the effect of strain on the easy axis direction as well as predicting 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. Finally, magnetic stiffness computed within the same model of ferromagnetism is presented, and is shown to explain the domain width in perpendicular films. It is emphasized that a rather large magnitude of the stiffness accounts for both the excellent micromagnetic properties of ferromagnetic (III, Mn)V films and the quantitative applicability of the mean-field approximation to these materials.

1. Introduction

In recent years a considerable effort has been devoted to understanding the nature of carrier-controlled ferromagnetism in tetrahedrally coordinated diluted magnetic semiconductors [1, 2]. It has been found that in these materials conceptual difficulties of charge transfer insulators and strongly correlated disordered metals are combined with intricate properties of heavily doped semiconductors, such as Anderson–Mott localization and defect creation by self-compensation mechanisms. Nevertheless, (Ga, Mn)As and related compounds are emerging as the best understood ferromagnets. In particular, the theory [3] built on Zener's model of ferromagnetism, the Ginzburg–Landau approach to the phase transitions, and the Kohn–Luttinger *kp* theory of semiconductors describes a number of pertinent material properties. Within this model and its variants, the magnitude of the Curie temperature T_C in Mn-doped GaAs, InAs, GaSb, and 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 localized spins are mediated by the holes in the weakly perturbed valence band [8]. This Zener/RKKY type of view is supported by *ab initio* computations within the density functional theory provided that self-interaction corrections or the Hubbard U are taken into account in the description of strongly correlated d electrons on Mn ions [9]. At the same time, the assumption that the relevant carriers reside in the p-like valence band makes it possible to describe various magneto-optical [4, 10] and magnetotransport properties of (Ga, Mn)As, including the anomalous Hall effect and anisotropy magnetoresistance [10, 11] as well the negative magnetoresistance caused by the orbital weak-localization effect [12]. Importantly, the understanding of (III, Mn)As alloys has provided a basis for the development of novel methods enabling magnetization manipulation and switching [13].

However, in the context of room-temperature semiconductor spintronics especially promising are the group III nitrides [14–16] and group II oxides [17, 18]. In these systems, the short bond length results in a strong hybridization between valence band states and open d shells of transition metal impurities, which is predicted [3, 4] to enhance the Curie temperature of the hole-mediated ferromagnetism. Indeed, spontaneous magnetization persisting to above room temperature has been detected for a number of magnetically doped nitrides and oxides. Surprisingly, however, the indications of ferromagnetism have often been found in n-type materials, while—owing to relatively small magnitudes of both s–d exchange coupling and density of states—the carrier-induced ferromagnetism can occur only under rather restricted conditions in n-type Mn-based DMSs [19, 20]. Thus, we are still at the beginning of the road to understanding and controlling doping, defects, and magnetism of these systems [21].

In this paper, a brief review of micromagnetic properties of (Ga, Mn)As and related compounds is given. Interestingly, despite much lower spin and carrier concentrations compared to ferromagnetic metals, these materials exhibit excellent micromagnetic characteristics, including well defined magnetic anisotropy and large and 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 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. Origin of magnetic anisotropy

As the energy of dipole–dipole magnetic interactions depends on the dipole distribution, there exists a so-called shape anisotropy. In particular, for thin films, the difference in energy density corresponding to the perpendicular and in-plane orientations of magnetization M is given by

$$E = \mu_0 M^2 / 2, \quad (1)$$

which leads to the anisotropy field $\mu_0 H_A = \mu_0 M$ of about 60 mT for $\text{Ga}_{0.95}\text{Mn}_{0.05}\text{As}$.

Already early studies of the ferromagnetic phase in (In, Mn)As [22] and (Ga, Mn)As [23] have demonstrated the existence of magnetic anisotropy, whose character and magnitude implied a sizable contribution of a microscopic origin. Magneto-crystalline anisotropy is usually associated with the interaction between spin and orbital degrees of freedom of the magnetic ion d electrons. According to the model advocated here, these electrons are in the d^5 configuration. For such a case the orbital momentum $L = 0$, so that effects stemming from the spin–orbit coupling are expected to be rather weak. It has, however, been noted

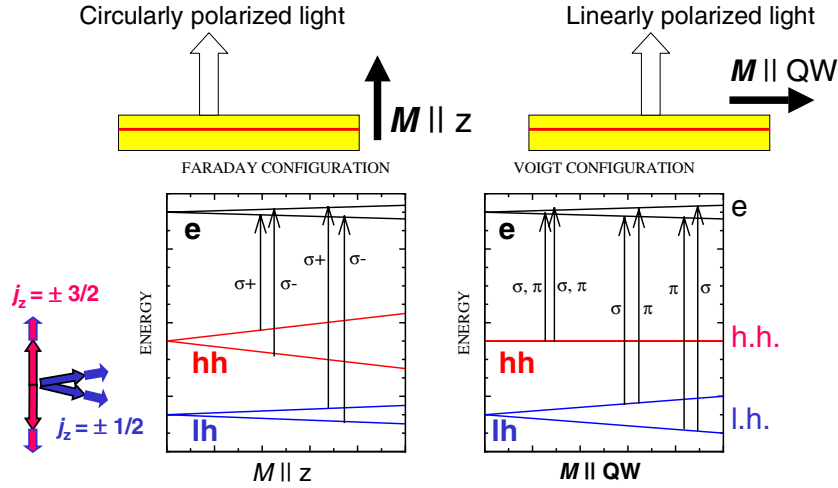


Figure 1. Illustration of subband splittings and optical transitions in a Mn-doped quantum well of tetrahedrally coordinated semiconductors for compressive strain and for two orientations of magnetization M in respect to growth direction Z (courtesy of Kossacki *et al* [25]).

(This figure is in colour only in the electronic version)

that the interaction between the localized 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, 24].

In order to visualize the origin, direction, and magnitude of the expected magnetic anisotropy it is instructive to refer to the electronic structure of the hole subbands in a Mn-doped quantum well of zinc-blende compounds, an issue familiar to the semiconductor community. Owing to a strong spin–orbit interaction, $s \parallel l$ in the case of the holes the $j = 3/2$ level resides 1 eV above the $j = 1/2$ level in tellurides. As shown in figure 1, in the presence of confinement the ground state subband assumes a heavy-hole character, for which $j_z = \pm 3/2$. Therefore, provided that only the ground state subband is occupied, the hole spins are oriented along the growth direction. Now, since the p–d exchange interaction has a scalar form, $H_{pd} \sim \mathbf{sS}$, the in-plane Mn spin magnetization M will not affect the heavy-hole subband. This means that perpendicular magnetic anisotropy is expected. Indeed, only for such a magnetization orientation can the holes lower their energy by the coupling to the Mn spins.

However, the energetic distance between the heavy-hole $j_z = \pm 3/2$ and light-hole $j_z = \pm 1/2$ subbands depends on strain. In particular, the ground state subband can acquire a *light-hole* character for a sufficiently large magnitude of the biaxial *tensile* strain. In the latter case, the in-plane component of the hole spin is greater than the perpendicular component, so that 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.

Recently, quantum wells of modulation-doped p-type (Cd, Mn)Te under compressive and tensile strain were grown by selecting appropriate substrates, (Zn, Cd)Te and CdTe, respectively [25]. Figure 2 shows the inverse energy splitting of the photoluminescence line as a function of temperatures for various orientations of the perpendicular and in-plane orientations of the weak external magnetic field [25]. As observed previously [26], the splitting occurs only for the perpendicular orientation for compressive strain, and points to the Curie temperature

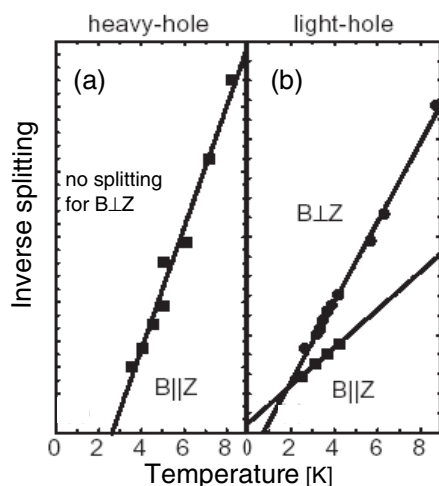


Figure 2. The Curie–Weiss behaviour above the Curie temperature obtained from photoluminescence measurements of p-(Cd, Mn)Te quantum wells under compressive (a) and tensile epitaxial strain (b) in the magnetic field parallel and perpendicular to the growth axis Z (points). The line splitting proportional to weak-field Mn magnetization is expressed in arbitrary units. The straight lines are drawn through experimental points (after Kossacki *et al* [25]).

of 2.7 K. A quite different behaviour, consistent however with the discussion presented above, is observed for the tensile strain. Here, the splitting is seen for the two orientations of the field. Furthermore, the in-plane direction of the easy axis is observed, as documented by the positive Curie–Weiss temperature for this orientation. The value of this temperature is only about 0.5 K, consistent with a weaker exchange splitting at given M for $j_z = \pm 1/2$ compared to $j_z = \pm 3/2$.

3. Strain-induced anisotropy and reorientation transition

A detailed theoretical analysis of anisotropy energies and anisotropy fields in films of (Ga, Mn)As has been carried out for a number of experimentally important cases within the p–d Zener model [4, 24]. In particular, the cubic anisotropy as well as uniaxial anisotropy under biaxial epitaxial strain have been examined as a function of the hole concentration p . Figure 3 shows the computed anisotropy field as a function of the hole concentration and Mn magnetization for (Ga, Mn)As films under compressive and tensile strains, which can be imposed by, for instance, GaAs and (In, Ga)As substrates, respectively [4]. Both shape and magneto-crystalline anisotropies have been taken into account. We see that in agreement with the discussion presented in the previous section, the perpendicular and in-plane orientation of the easy axis is expected for the compressive and tensile strain, respectively, provided that the hole concentration is sufficiently small. However, according to theory, a reorientation of the easy axis direction is expected at higher hole concentrations. Furthermore, in a certain concentration range the character of magnetic anisotropy is computed to depend on the magnitude of spontaneous magnetization, that is on the temperature.

The predicted reorientation transition has been detected in (In, Mn)As [27], (Al, Ga, Mn)As [28], and (Ga, Mn)As films [29, 30]. Figure 4 depicts the pertinent experimental results for (Ga, Mn)As with 5.3% of Mn deposited onto GaAs, which shows the in-plane and perpendicular components of spontaneous magnetization measured as a function

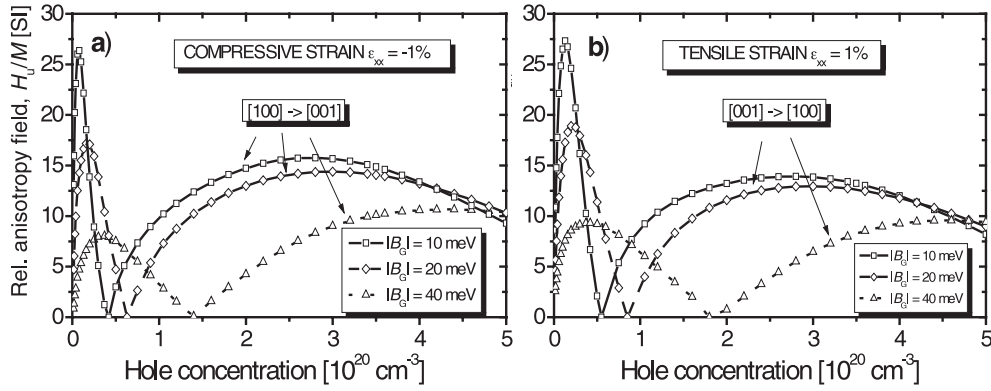


Figure 3. Computed minimum magnetic field H_u (divided by M) necessary to align magnetization M along the hard axis for compressive (a) and tensile biaxial strain (b) in (Ga, Mn)As films at various values of the spin-splitting parameter B_G . For compressive strain, the easy axis is along the [001] direction and in the (001) plane at low and high hole concentrations, respectively. The opposite behaviour is observed for tensile strain. The symbol [100] \rightarrow [001] means that the easy axis is along [100], so that H is applied along [001]. The value of $B_G = 30$ meV corresponds to the saturation value of M for $\text{Ga}_{0.95}\text{Mn}_{0.05}\text{As}$ (after Dietl *et al* [4]).

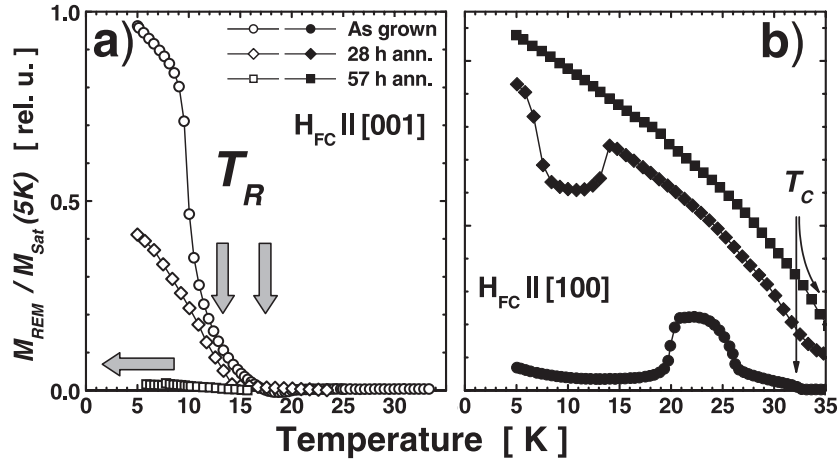


Figure 4. Temperature dependence of the remanent magnetization as measured in perpendicular [001] (a) and in-plane [100] (b) configurations for a $\text{Ga}_{0.947}\text{Mn}_{0.053}\text{As}$ sample prior to annealing (circles) and after annealing (diamonds and squares). The sample is cooled down through T_C in the field $H_{FC} = 0.1$ T, which is higher at least by a factor of ten than the coercive field H_c . Then, the field is removed at 5 K, and the measurement of the magnetization component M along the direction of H_{FC} commences on increasing temperature in the residual field $H_r < 6 \mu\text{T}$. 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_R when the cross-over to in-plane magnetic anisotropy takes place (after Sawicki *et al* [30]).

of temperature for samples with various hole concentrations changed by low-temperature annealing [30]. A flipping of the easy axis direction is clearly seen. The computed phase diagram for Mn concentration x and compressive strain ϵ_{xx} in question compared to the experimental results for this film is shown in figure 5. In view that theory is developed with no adjustable parameters the agreement between experimental and computed concentrations

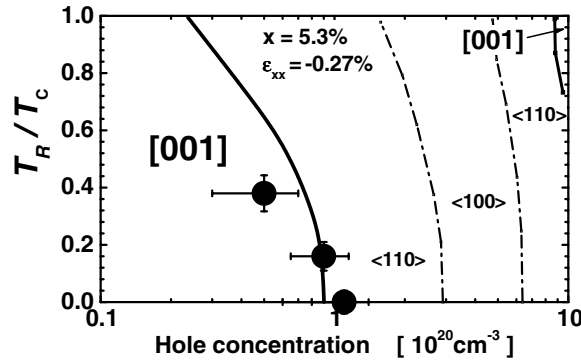


Figure 5. Experimental (full points, taken from figure 4) and computed values (thick curves) of the ratio of the reorientation to Curie temperature for the perpendicular to in-plane magnetic anisotropy transition. Dashed curves mark expected temperatures for the reorientation of the easy axis between $\langle 100 \rangle$ and $\langle 110 \rangle$ in-plane directions (after Sawicki *et al* [30]).

and temperature corresponding to the reorientation transition is very good. Furthermore, according to figure 3, the predicted anisotropy field in the limit of large carrier concentrations $\mu_0 H_u \approx 10 \mu_0 M \approx 6$ T for $x = 0.05$ and $|\varepsilon_{xx}| = 1\%$, after rescaling to the actual magnitudes of $|\varepsilon_{xx}|$, H_u is consistent with the available findings for both compressive and tensile strain [26, 31, 36].

4. In-plane magnetic anisotropy

According to the discussion above, the easy axis assumes the in-plane orientation for typical carrier concentrations in the most thoroughly studied system (Ga, Mn)As/GaAs. In this case, according to the theoretical predictions presented in figure 5 as well in figure 9 of [4] and in figure 6 of [24] the easy axis is expected to switch between $\langle 100 \rangle$ and $\langle 110 \rangle$ in-plane cubic directions as a function of p . Surprisingly, however, only the $\langle 100 \rangle$ biaxial magnetic symmetry has so far been observed [30–37] in films of (Ga, Mn)As/GaAs at low temperatures. Nevertheless, the corresponding in-plane anisotropy field assumes the expected magnitude, of the order of 0.1 T, which is typically much smaller than that corresponding to the strain-induced energy of magnetic anisotropy. It is possible that anisotropy of the hole magnetic moment, neglected in the theoretical calculations [4, 24], stabilizes the $\langle 100 \rangle$ orientation of the easy axis. However, whether such a model will explain simultaneously the recently reported $\langle 110 \rangle$ biaxial symmetry in (In, Mn)As/(In, Al)As films [38] remains to be shown.

In addition to the cubic in-plane anisotropy, the accumulated data for both (Ga, Mn)As/GaAs [30–37] and (In, Mn)As/(In, Al)As point to a non-equivalence of $[110]$ and $[\bar{1}10]$ directions, which leads to the in-plane uniaxial magnetic anisotropy. As shown in figure 6 [30], remanent magnetization M measured along the $[\bar{1}10]$ direction vanishes completely above 15 K, indicating that this is the hard direction in this film. We also note that when $M_{[\bar{1}10]}$ 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 [37], which—in particular—rules out the effect of Mn oxide accumulated at the free surface [39, 40]. Thus, the observed in-plane anisotropy points to a puzzling symmetry breaking in the film body.

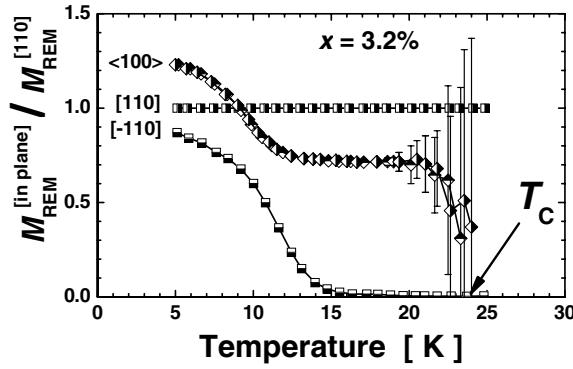


Figure 6. Experimental evidence for the uniaxial anisotropy along the [110] direction in $\text{Ga}_{0.97}\text{Mn}_{0.03}\text{As}$ film. The magnetic remanence is measured for four major in-plane directions and its magnitude is normalized by the data of the [110] case. Note that the sudden drop of M along $[\bar{1}10]$ at $T < T_C$ may wrongly indicate too low a value of T_C , if only this orientation is probed (after Sawicki *et al* [30]).

5. Magnetic stiffness and domain structure

Another important characteristic of any ferromagnetic system is magnetic stiffness A , which describes the energy penalty associated with the local twisting of the direction of magnetization. Remarkably, A determines the magnitude and character of thermodynamic fluctuations of magnetization, the spectrum of spin excitations as well as the width and energy of domain walls. An important result is that the magnetic stiffness computed within the 6×6 Luttinger model is greater almost by a factor of 10 than that expected for a simple spin degenerate band with the heavy-hole band-edge mass [41]. This enhancement, which strongly stabilizes the spatially uniform spin ordering, stems presumably from p-like symmetry of the valence band wavefunctions, as for such a case the carrier susceptibility (the Lindhard function) decreases strongly with q [42].

The structure of magnetic domains in (Ga, Mn)As under tensile strain has been determined by micro-Hall probe imaging [43]. The regions with magnetization oriented along the $[001]$ and $[00\bar{1}]$ easy axes form alternating stripes extending in the [110] direction. As shown in figure 7, the experimentally determined stripe width is $W = 1.5 \mu\text{m}$ at 5 K for a $0.2 \mu\text{m}$ film of $\text{Ga}_{0.957}\text{Mn}_{0.043}\text{As}$ on $\text{Ga}_{0.84}\text{In}_{0.16}\text{As}$, for which tensile strain of $\varepsilon_{xx} = 0.9\%$ is expected. According to micromagnetic theory, W is determined by the ratio of the domain wall energy to the stray field energy (1), $\lambda_c = 4(AH_u/2)^{1/2}/\mu_0 M^3/2d$, where d is film thickness. As shown in figure 7, the computed value with no adjustable parameters $W = 1.1 \mu\text{m}$ [44] compares favourably with the experimental finding, $W = 1.5 \mu\text{m}$ at low temperatures. However, the model predicts much weaker temperature dependence of W than that observed experimentally, which was linked [44] to critical fluctuations, disregarded in the mean-field approach.

Recently, a current-induced domain-wall motion in a (Ga, Mn)As film under tensile strain has been demonstrated by the magneto-optical Kerr effect [45]. It may appear surprising that a domain free uniform magnetization was observed in the $20 \times 20 \mu\text{m}^2$ active region of the device despite the perpendicular orientation of the easy axis. It turns out, however, for the actual film thickness of 25 nm, that the expected [44] stripe domain width W is of the order of a centimetre. Magneto-optical effects [35] and a scanning Hall microscope [46] have also been employed to visualize the existence of large and regular domains in good quality (Ga, Mn)As films with the in-plane easy axis.

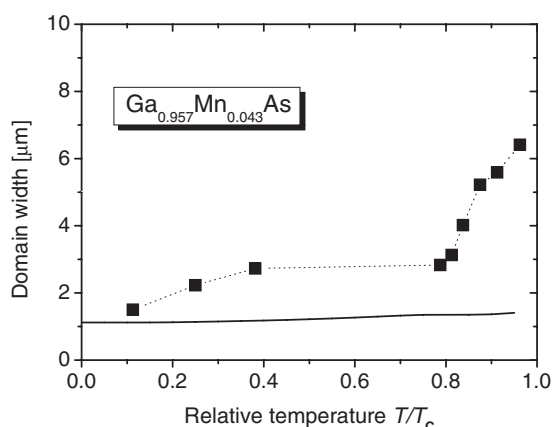


Figure 7. Temperature dependence of the width of domain stripes as measured by Shono *et al* [21] for the $\text{Ga}_{0.957}\text{Mn}_{0.043}\text{As}$ film with the easy axis along the growth direction (full squares). The computed domain width is shown by the solid line (after Dietl *et al* [44]).

6. Conclusions

In summary, experimental and theoretical 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 theory [4, 24] these reflect spin anisotropy of the valence band subbands whose shape depends on strain, while the splitting and population on magnetization and hole concentration. Excellent micromagnetic properties of these materials can be linked to a large magnitude of the magnetic stiffness, which diminishes the role of magnetization fluctuations and domain formation. Such a picture is, presumably, altered in the case of p-type II–VI ferromagnetic semiconductors, where the competing short-range but strong antiferromagnetic interactions may affect micromagnetic characteristics.

Acknowledgments

I would like to thank my numerous co-workers, as indicated in the reference list, for many years of fruitful and enjoyable interactions. This work was partly supported by the State Committee for Scientific Research as well as by the FENIKS project (EC:G5RD-CT-2001-0535).

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