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Magnetic domains in III-V magnetic semiconductors

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Recent progress in the theoretical understanding of magnetic anisotropy and stiffness in III-V magnetic semiconductors is exploited for predictions of magnetic domain characteristics and methods of their tuning. We evaluate the width and the energy of domain walls as well as the period of stripe domains in perpendicular films. The computed stripe width $W=1.1$ μm for $Ga_{0.957}Mn_{0.043}As/In_{0.16}Ga_{0.84}As$ compares favorably to the experimental value 1.5 μm, as determined by Shono *et al.* [Appl. Phys. Lett. **77**, 1363 (2000)].

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The utility foreseen for ferromagnetic semiconductors rests on the possibility of tailoring their electronic and magnetic properties on the same footing. Indeed, successful control of the Curie temperature T_C by hole density has been achieved in Mn-based IV-VI,[1] III-V,[2] and II-VI (Refs. 3 and 4) semiconductor compounds. It has also been found that both biaxial strain imposed by lattice mismatch[5] and confinement in quantum structures[3] can serve to engineer the direction of the easy axis. Furthermore, light-induced changes of the magnetic phase in[6] (In,Mn)As/(Al,Ga)Sb and[3] (Cd,Mn)Te/(Cd,Zn,Mg)Te heterostructures has been demonstrated. Finally, the possibility to tune T_C of (In,Mn)As quantum wells by metallic gates has also been shown.[7] We can anticipate many demonstrations of tunable magnetic and magnetotransport properties in the future.

These tuning capabilities along with T_C's as high as[8] 110 K in (Ga,Mn)As with 5.3% Mn have triggered a considerable theoretical effort to elucidate the origin of ferromagnetism in III-V magnetic semiconductors. While there is general agreement that the Mn constituent introduces both localized spins and itinerant holes, the nature and energy of the Mn-derived states, the role of intrinsic defects, the relative importance of charge and spin fluctuations, as well as the consequences of electrostatic and magnetic disorder are still under debate.[9]

Recently, a quantitative theory of hole-mediated ferromagnetism in tetrahedrally coordinated magnetic semiconductors has been put forward by the independent effort of two teams consisting of the present authors and co-workers.[10–13] In this theory, the ferromagnetic interaction between spins localized on the d shells of the magnetic ions is mediated by holes in the valence band. The free energy of the hole liquid is computed by diagonalizing the 6×6 Luttinger Hamiltonian, which contains $k\cdot p$, spin-orbit, and p-d exchange interactions, the latter taken into account in the molecular-field and virtual-crystal approximations. The influence of electrostatic and spin disorder on magnetic properties is neglected, since they are not expected to have a qualitative impact on spin-polarized band electron thermodynamic properties. Hole-hole interactions can be taken into account in the spirit of Fermi-liquid theory. The use of a mean-field approximation for the coupled band-electron and local-moment systems can be justified by the long-range character of the carrier-mediated spin-spin interaction, at least when the ratio of hole to Mn density is small and the Fermi energy is large.[14] It has been shown[12] that this model, with material parameters known from independent experiments, satisfactorily explains the magnitude of T_C, the temperature dependence of the spontaneous magnetization, the strength and strain dependence of the magnetic anisotropy, as well as the spectral dependence of the magnetic circular dichroism in (Ga,Mn)As. The hole-spin polarization has also been evaluated.[12] To address spin fluctuations a theoretical description of ferromagnetism beyond the mean-field approximation has been developed,[15] from which the magnon excitation spectrum,[13,15] and the magnetic stiffness could be deduced.[13]

In this Rapid Communication, we exploit this progress in the theoretical description of magnetic anisotropy[10–13] and magnetic stiffness[13] to address the domain structure in epitaxial layers of (Ga,Mn)As. The comparison between the computed and experimentally observed width of domain stripes presented here constitutes an additional test of current theory. Moreover, the determined values for anisotropy energy, domain-wall energy and width may serve for optimizing the design of, for instance, spin injection,[16] spin tunneling,[17,18] or micromechanical nanostructures[19] of (Ga,Mn)As.

For the calculation presented below we adopt band-structure parameters, elastic constants, and deformation potentials of GaAs, a set of values employed in our previous works[10–13] for (Ga,Mn)As. The Mn ions are assumed to be in the d^5 configuration, i.e., their spin is $S=5/2$ and the Mn Landé factor $g=2.0$. For the p-d exchange energy we take[10,12] $\beta N_o=-1.2$ eV, which for the cation concentration of GaAs, $N_o=2.21\times10^{22}$ cm^{-3}, corresponds to $J_{pd}\equiv-\beta=0.054$ eV nm^3. The Fermi-liquid parameter $A_F=1.2$ enters

the enhancement of T_C and of the p-d exchange splitting $\Delta = A_F J_{pd} M/(g\mu_B)$ of the valence band at magnetization M of the Mn spins.[12]

Another important parameter characterizing epitaxial layers is the magnitude of biaxial strain. It depends on the layer thickness d and the difference between the lattice parameters of the substrate and the layer, $\Delta a = a_s - a(x)$. In the case of (Ga,Mn)As that is obtained by low-temperature MBE, films with d as large as 2 μm are not relaxed.[20] For such layers, the relevant components of the strain tensor assume the form $\epsilon_{xx} = \epsilon_{yy} = \Delta a/a$ and $\epsilon_{zz} = -2\epsilon_{xx} c_{12}/c_{11}$, where the c_{ij} are elastic constants. Since $d\,a(x)/d\,x = 0.032$ nm,[5] the appropriately thin layer of (Ga,Mn)As deposited on GaAs or (Al,Ga)As is under compressive strain but the use of (Ga,In)As substrates can result in a tensile strain.

It has been demonstrated by Ohno *et al.*[5] that for films under compressive (tensile) strain the easy axis is in plane (perpendicular to film growth direction). Quantitatively, $\epsilon_{xx} = -0.2\%$ for the $Ga_{0.965}Mn_{0.035}As$ film on GaAs, for which Ohno *et al.*[5] determined the magnitude of the magnetic field H_a aligning magnetization along the hard axis. Tensile strain of $\epsilon_{xx} = 0.90\%$ is expected for the 0.2 μm perpendicular film of $Ga_{0.957}Mn_{0.043}As$ on $Ga_{0.84}In_{0.16}As$, a sample employed by Shono *et al.*[21] to examine the domain structure. Importantly, the theory referred to above[10–12] reproduces correctly the strain dependence of magnetic anisotropy and, in particular, explains the magnitude of H_a.[12] Furthermore, it has been found[11,12] that for the relevant values of strain, hole concentrations p, and magnitudes of magnetization M, the energy density K_u that characterizes uniaxial magnetic anisotropy is greater than the corresponding cubic anisotropy terms K_c and the energy density of the stray fields $K_d = \mu_o M^2/2$. Thus, (Ga,Mn)As can be classified as a uniaxial ferromagnet.

We discuss the domain structure in terms of micromagnetic theory.[22,23] Within this approach, a uniaxial ferromagnet is characterized by the anisotropy energy K_u, the magnetic stiffness A, and the saturation magnetization M_s. The procedure we adopt here consists of evaluating the temperature-dependent magnetization $M(T)$, and thus the exchange splitting $\Delta(T)$, in mean-field approximation. Then, the magnetic anisotropy $K_u(\Delta)$ and the spin stiffness $A(\Delta)$ are calculated. We believe that this procedure is well grounded at low temperatures. However fluctuation corrections will be more important at nonzero temperatures, particularly in the critical region near the Curie temperature T_C.

An important question arises whether the continuous-medium approximation underlying the micromagnetic theory is valid in diluted magnetic semiconductors, which contain a relatively low concentration of magnetic ions and an even lower concentration of carriers. To address this question we note that the shortest length scale of micromagnetic theory is the width of the Bloch domain wall,

$$\delta_W = \pi\sqrt{A/K_u}. \tag{1}$$

This length has to be compared to the mean distance between holes, $r_m = \sqrt[3]{3/(4\pi p)}$. For concreteness, we consider $Ga_{0.957}Mn_{0.043}As$ on $Ga_{0.84}In_{0.16}As$ for which $T_C = 80$ K.[21] Assuming a hole concentration $p = 3\times10^{20}$ cm^{-3} we obtain the mean-field Curie temperature $T_C = 91$ K. The actual value of the hole concentration is uncertain; the mean-field T_C would be 80 K for $p = 2.5\times10^{20}$ cm^{-3}. Since the Fermi energy is greater than the exchange splitting Δ at low temperatures (this corresponds to the "weak-coupling" or "RKKY" regime[24]) the redistribution of holes between the four valence subbands is only partial, and both the spin stiffness $A(\Delta)$ and anisotropy constant $K_u(\Delta)$ are proportional to Δ^2, except for large splittings Δ, where the increase of the anisotropy K_u with Δ is somewhat weaker. Hence, the ratio A/K_u and thus the domain-wall width δ_W are virtually independent of Δ, that is of temperature. For instance, for $p = 3\times10^{20}$ cm^{-3}, the low-temperature domain-wall width is $\delta_W = 14.9$ nm and decreases to 14.6 nm in the opposite limit $T \to T_C$. Thus, the value determined for $\delta_W \approx 15$ nm is by more than a factor of ten longer than the mean hole distance r_m. We checked that $\delta_W \gg r_m$ in the whole relevant range of the hole concentrations down to 10^{20} cm^{-3}. We conclude that the micromagnetic theory in its standard continuous-medium form is suitable for modeling the domain structure in (Ga,Mn)As.

In the case of perpendicular easy-axis films, a competition between energies associated with the stray fields and the formation of the Bloch domain walls results in a simple stripe domain structure in the demagnetized thermal equilibrium state.[25] Indeed, Shono *et al.* observed such a structure in $Ga_{0.957}Mn_{0.043}As/Ga_{0.84}In_{0.16}As$ by means of a micro-Hall scanning probe.[21] In particular, well-defined stripes of width W increasing from 1.5 μm at 9 K to 2.5 μm at 30 K were observed. Above 60 K, the stripes were less regular. Their width was evaluated to change from 3 to 6 μm in the temperature range between 65 and 77 K. The stripes were oriented along [110] at low temperatures but tended to lay along [100] above 60 K.

To interpret the experimental results we recall[22,25] that for film width d much larger than domain-wall width δ_W and $K_u \gg K_d$, the stripe width W is determined by a solution of the transcendental equation,

$$\lambda_c = (P^2/\pi^3) \sum_{n=1,3,5,\ldots} n^{-3}[1-(1+2\pi n/P)e^{-2\pi n/P}]. \tag{2}$$

Here, P is the normalized stripe period $P = 2W/d$ and the parameter λ_c describes the ratio of the Bloch domain-wall energy $\gamma_W = 4\sqrt{AK_u}$ and the stray-field energy K_d,

$$\lambda_c = 4\sqrt{AK_u}/(\mu_o M^2 d). \tag{3}$$

Figure 1 shows λ_c as a function of T/T_C computed for the film in question. Again the dependence on temperature is weak as both the denominator and numerator are to a good accuracy proportional to Δ^2.

We now evaluate the domain width $W(T)$. The results and a comparison to the experimental data of Shono *et al.*[21] are presented in Fig. 2. We see that the computed value for low temperatures, $W = 1.1$ μm, compares favorably with the experimental finding, $W = 1.5$ μm. The difference between the

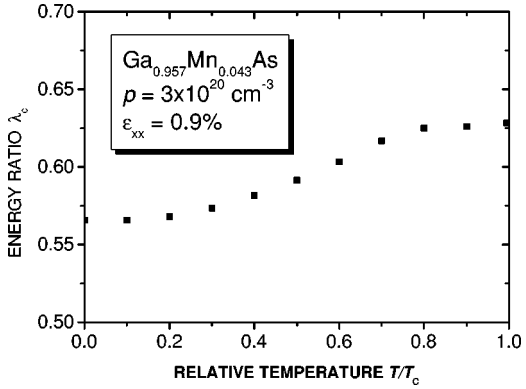

FIG. 1. Computed ratio of domain wall to stray-field energy, λ_c [Eq. (3)] as a function of reduced temperature for a $Ga_{1-x}Mn_xAs$ film with the easy axis along the growth direction.

two may stem from the uncertainty in the input parameters, as the results calculated for a value of λ_c 1.8 times larger point to a strong sensitivity of the theoretical results to the parameter values. Moreover, recent Monte Carlo simulations[26] demonstrate that disorder in positions of magnetic ions tends to enhance the stiffness *A*, and thus the domain width *W*. Much below T_C, a rather weak temperature dependence of *W* is observed experimentally, a behavior consistent with the theoretical expectations. At the same time, it is clear from Fig. 2 that a strong temperature dependence of *W* at higher temperatures is not explained by our model. It seems likely that the break in the temperature dependence of the domain width $W(T)$ around 60 K marks the beginning of the critical regime. In this regime, long-length-scale fluctuations in the magnetization *M*, not accounted for in our theory, become important. These will have a larger effect on the stray-field energy K_d, which is sensitive to fluctuations on scales shorter than the domain width *W*, than on the domain-wall energy γ_W, which is sensitive only to fluctuations on length scales shorter than the domain-wall width δ_W.

Next, we comment on the crystallographic orientation of the domain stripes. The computed direction of the in-plane

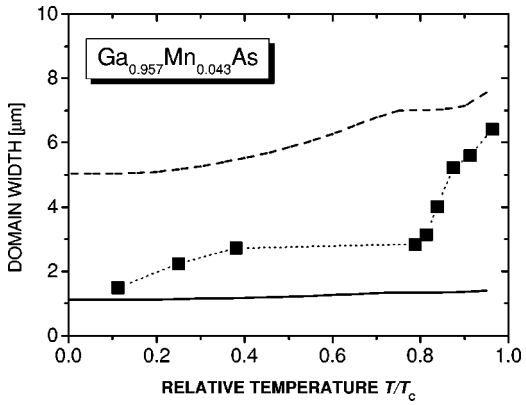

FIG. 2. Temperature dependence of the width of domain stripes as measured by Shono *et al.*[21] for the $Ga_{0.957}Mn_{0.043}As$ film with the easy axis along the growth direction (full squares). Computed domain width for the parameter λ_c depicted in Fig. 1 is shown by the solid line. The dashed line is computed assuming that λ_c is by a factor of 1.8 greater.

easy axis oscillates between [100] and [110] directions as a function of the hole concentration *p* and the splitting Δ.[11,12] For the film in question we find that the lowest energy will have domain walls laying along [100] or equivalent directions, provided that the hole concentration *p* is greater than 2.5×10^{20} cm^{-3}. This stripe direction is in disagreement with the experimental results[21] described above. However, for slightly smaller values of *p*, the predicted behavior is in accord with the findings. In particular, for $p=2\times10^{20}$ cm^{-3}, the wall direction is expected to assume [110] or equivalent directions for large exchange splittings $\Delta\rightarrow\Delta_s$, that is at low temperatures, but to extend along [100] or equivalent directions at higher temperature, $\Delta<\Delta_s/2$.

It is well known that the domain structure of perpendicular easy-axis films exhibits an interesting evolution as a function of the magnetic field along the easy axis. When the field increases, stripes magnetized along the field grow, while those antiparallel to the field shrink. However, beyond a critical field value, cylindrical bubbles rather than stripes have lower energy. It would be interesting to search for such domains in perpendicular films of III-V magnetic semiconductors. At the same time, history-dependent metastable domain arrangements are expected to develop, for instance, a ''froth'' structure in the remanent state. The magnetic field H_{cb} at which domains vanish entirely under conditions of thermal equilibrium increases with $1/\lambda_c$, so that $H_{cb}\rightarrow M_s$ for $\lambda_c\ll1$. For the case under consideration, $\lambda_c\approx0.5$ according to Fig. 1, which corresponds to $\mu_oH_{cb}\approx0.1\mu_oM_s\approx5.5$ mT. This is consistent with the observed coercive force $\mu_oH_c\approx20$ mT and square hysteresis for such a film.[20] Actually, the fact that $H_c>H_{cb}$ implies the existence of a domain pinning mechanism.

In view of the interest in magnetic nanocrystals, the length scale below which a ferromagnetic sample is in a single-domain state, is an interesting material parameter. Such a length scale is shape dependent. We consider square samples of ferromagnetic films with dimensions $W\times W\times d$. In thermodynamic equilibrium, the width W_{SD} below which the material is in a single-domain state is determined by the film thickness *d* and $l_c=\gamma_W/2K_d=\lambda_c d$. For the perpendicular film discussed above, $d=0.2$ μm and $l_c=0.1$ μm. For these values, the single-domain width is, according to numerical results of Hubert and Schäfer,[22] $W_{SD}=1.2$ μm. For such small single-domain particles, the celebrated Stoner-Wohlfarth theory predicts an abrupt switching of the magnetization direction in a magnetic field along the easy axis at $H_f=H_a\equiv2K_u/(\mu_oM_s)$. Thus, in this case the coercive force is equal to the anisotropy field aligning magnetization along the hard axis, $\mu_oH_c=\mu_oH_a=670$ mT for the material in question. It would be interesting to check experimentally the actual magnitude of H_c in nanostructures of III-V DMS. On the other hand, it is known already that in the case of macroscopic films $H_c\ll H_a$, reflecting the inevitability of domain nucleation processes, associated for instance with space fluctuations of H_a. The appearance of reverse domains followed by domain wall motion results in the complete reversal of magnetization at H_c in the range $H_a>H_c>H_{cb}$. The wall motion begins when the field-induced torque on the wall magnetization overcomes the wall pinning force.

In conclusion, our results imply that despite relatively small concentrations of magnetic ions and carriers, domain properties of III-V DMS are, in many respects, similar to those of standard ferromagnets. In particular, domain characteristics can be described in terms of micromagnetic theory. Such an approach, combined with our microscopic theory of hole-mediated ferromagnetism, predicts the direction and strength of magnetic anisotropy as well as characteristic dimensions of the domains correctly. We note that the values predicted for both anisotropy energies and domain-wall energies are qualitatively dependent on an accurate representation of the host semiconductor valence band. Further experimental studies on both macroscopic films and nanostructures with fine spatial and time resolution will certainly improve our understanding of this ferromagnetic system, opening the doors wider for domain engineering. Furthermore, the role of the magnetostriction and, in particular, its contribution to the strain tensor remains to be elucidated. On the theoretical side, it will be interesting to see to what extent intrinsic fluctuations in the Mn distribution, the distribution of other extrinsic defects, and the carrier density distributions they produce, account for domain nucleation and pinning fields.

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