

High-field magnetocrystalline anisotropic resistance effect in (Ga,Mn)As

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As the magnetization rotates in the (001) plane of epitaxial (Ga,Mn)As films, we observe both two- and fourfold oscillations of comparable magnitude in the longitudinal resistivity. This behavior is different from the usual anisotropic magnetoresistance effect in polycrystalline films. The fourfold or cubic symmetry vanishes at the Curie temperature T_C , indicating that it originates from the long-range ferromagnetic phase in single crystal films. In contrast, the twofold symmetry persists above T_C , suggesting its origin to be from the alignment of spins with random orientations. However, the transverse, or planar Hall, resistivity only contains a twofold oscillation. The temperature dependence of the magnetocrystalline anisotropic resistance effect is explained by a two-component model.

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The anisotropic magnetoresistance (AMR) effect in ferromagnetic (FM) materials arises from the difference in the scattering rate when the magnetization direction is oriented parallel or perpendicular to an electrical current.¹ It is a convenient tool often used for characterizing the state of the magnetization in ferromagnetic metals² and semiconductors.^{3,4} In ferromagnetic semiconductors such as GaMnAs, AMR has been linked to the intrinsic band structure properties in the presence of spin-orbit interaction.⁵ For polycrystalline materials, the resistivity typically shows a twofold symmetry because the magnetocrystalline effect in the randomly oriented grains is averaged out. In single crystal films, however, also due to the spin-orbit interaction, AMR may contain higher-order terms that reflect the symmetry of the crystals.⁶ Nevertheless, in many cases, the high-order terms are small and often neglected.^{7,8} Here, we have conducted a series of experiments in epitaxially grown GaMnAs films and found a very strong fourfold magnetoresistance effect that is correlated with the crystalline symmetry of the films. This fourfold term is found to be connected to the long-range FM order below T_C ; therefore, its temperature and field dependences reveal valuable information about the ferromagnetic ordering in the materials.

We studied two $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ samples with $x=0.039$ and 0.059 which were grown on GaAs(001) wafers at 250°C by molecular beam epitaxy in an As-rich environment. The samples were patterned into the standard Hall bar using photolithography and wet chemical etching. For resistivity measurements, the current flows along a [110] direction, as schematically shown in Fig. 1(a), and both the longitudinal and transverse or the planar Hall dc resistivities ρ_{xx} and ρ_{xy} are measured simultaneously using the four-probe method. The measurements are performed in a superconducting magnet equipped with a rotating sample holder, which allows us to continuously change the angle ϕ between the magnetic field H and the electric current I , as illustrated in Fig. 1(a). As the sample is rotated around [001], H always lies in the sample plane, and both ρ_{xx} and ρ_{xy} are recorded as angle ϕ is varied. The angular dependence is taken for different magnetic field

strengths and temperatures. Since both samples show similar behaviors in their angular dependence, we will focus our discussions on the $x=0.059$ sample below.

ρ_{xx} as a function of temperature at $H=0$ for $\text{Ga}_{0.941}\text{Mn}_{0.059}\text{As}$ shows a peak due to the spin disorder scattering enhanced resistivity,⁹ as shown in the inset of Fig. 1(b). From ρ_{xx} vs T , we can estimate the Curie temperature, $T_C \sim 49$ K, which agrees with the T_C found from the superconducting quantum interference device measurements. Figure 1(b) shows ρ_{xx} as a function of H for the same sample at

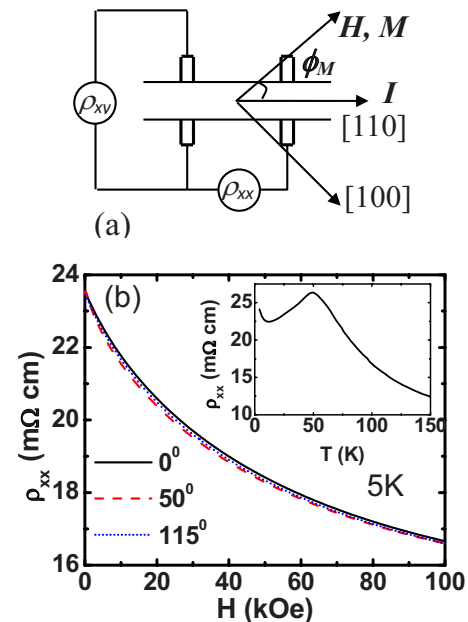


FIG. 1. (Color online) (a) Sketch of experimental geometry. Current I flows along [110] and magnetization M is aligned with the external field H which rotates in the (001) plane. (b) Longitudinal resistivity as a function of in-plane field at different field orientations, $\phi=0^\circ$, 50° , and 115° , at 5 K for $\text{Ga}_{0.941}\text{Mn}_{0.059}\text{As}$. The inset shows the resistivity as a function of temperature at zero field.

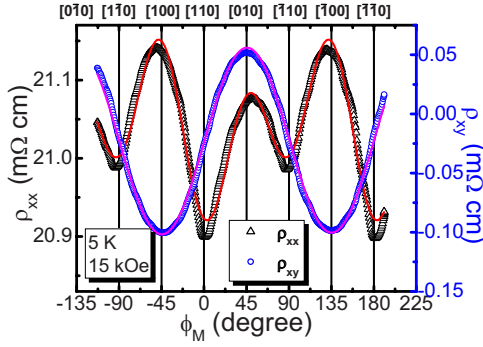


FIG. 2. (Color online) Angular dependence of the longitudinal resistivity ρ_{xx} and planar Hall resistivity ρ_{xy} at $H=15$ kOe and $T=5$ K for $\text{Ga}_{0.941}\text{Mn}_{0.059}\text{As}$. The solid lines are the fits.

5 K with H applied at three different angles with current I . In this field range, ρ_{xx} exhibits negative magnetoresistance (MR), which is in agreement with what was reported previously.¹⁰ This negative MR has been attributed to the scattering of carriers off paramagnetic Mn^{2+} ions or magnetic boundary polarons,^{11,12} or attributed to the weak localization effect.¹³ Since H far exceeds the coercive field H_c (<50 Oe at 5 K) or the anisotropy field H_k (~ 1000 Oe), the FM magnetization \mathbf{M} should be fully aligned with H . As seen in Fig. 1(b), this high-field MR actually depends on the direction of \mathbf{H} or \mathbf{M} .

The anisotropy of this high-field MR can be clearly seen as the sample is continuously rotated in a constant in-plane field whose strength is greater than that of the anisotropy field. At such a high field, ϕ_M , the angle between \mathbf{M} and I , should be nearly identical to ϕ , the angle between \mathbf{H} and I . In Fig. 2, we plot both ρ_{xx} and ρ_{xy} at 5 K with $H=15$ kOe. Both ρ_{xx} and ρ_{xy} oscillate as the sample is rotated about $[001]$, but the oscillation periods are different. Apparently, the oscillations in ρ_{xx} cannot be described by a simple $\cos 2\phi_M$ function that is normally expected for polycrystalline FM films. There is a large fourfold or cubic component present which peaks at approximately -45° , 45° , and 135° in ϕ_M . In GaMnAs (001) epitaxial films, this cubic symmetry simply coincides with the fourfold symmetry of the (001) plane of the zinc-blende crystalline structure. To include both types of symmetry, we use $\rho_{xx}=\rho_0(1+A_u \cos 2\phi_M + A_c \cos 4\phi_M)$ to fit our data and obtain the relative oscillation amplitudes for $\cos 2\phi_M$ and $\cos 4\phi_M$ terms, i.e., $A_u=0.25\%$ and $A_c=0.37\%$, respectively. The fourfold anisotropy is actually stronger than the twofold anisotropy in this case. By the conventional definition $\text{AMR}_{\text{ip}}\equiv[\rho_{J\parallel M} - \rho_{J\perp M}]/\rho_{J\perp M}$, the in-plane AMR ratio is calculated to be about -0.41% , and the sign agrees with the theoretical prediction.^{5,14} Unlike the symmetry in ρ_{xx} , the planar Hall resistivity ρ_{xy} only contains a twofold oscillation, as shown in Fig. 2, and it can be fitted well with a single $\sin 2\phi_M$ function. Note that ρ_{xy} has a nonzero offset, which is likely caused by a small ρ_{xx} component due to imperfect alignment between the two transverse leads.

For polycrystalline or isotropic ferromagnetic materials, ρ_{xx} and ρ_{xy} both depend on the relative angle ϕ_M between magnetization \mathbf{M} and current I . The AMR in single domain model can be described by two equations:¹

$$\rho_{xx} = \rho_{\perp} + (\rho_{\parallel} - \rho_{\perp})\cos^2 \phi_M, \quad (1)$$

$$\rho_{xy} = (\rho_{\parallel} - \rho_{\perp})\sin \phi_M \cos \phi_M, \quad (2)$$

where ρ_{\perp} and ρ_{\parallel} are the resistivities for $M \perp I$ and $M \parallel I$, respectively. Both ρ_{xx} and ρ_{xy} have a twofold or uniaxial symmetry about the current direction with the rotation of M and are intimately related to each other. The equations have been adopted to explain AMR observed in GaMnAs films.^{7,15,16} However, this picture is not applicable here since there is an additional strong fourfold term in ρ_{xx} . To see how different anisotropy terms contribute to ρ_{xx} and ρ_{xy} in single crystal films, here, we use a phenomenological description for the anisotropic transport. We assume that the resistivity contains up to the fourth order terms in direction cosines α

$$\rho_{ij} = a_{ij} + a_{kij}\alpha_k + a_{klj}\alpha_k\alpha_l + a_{klmij}\alpha_k\alpha_l\alpha_m + a_{klmni}\alpha_k\alpha_l\alpha_m\alpha_n + \dots, \quad (3)$$

where α is the direction cosines of \mathbf{M} with the cubic axes, and a_{ij} , a_{kij} , a_{klmij} , and a_{klmni} are the elements of the resistivity tensor of various orders. i and j can be in any of the three orthogonal directions, but in the context of our discussions, they are either along or perpendicular to the current in the film plane. The resistivity tensor ρ_{ij} then relates to AMR effect and the planar Hall effect through Ohm's law: $E_i = \rho_{ij}(\alpha)J_j$, where E_i is the electric field along i and J_j is the current density along j . For instance, if a current J_x flows along the x axis, the electric field E_x in this direction depends on the direction cosines of \mathbf{M} with respect to the cubic axes via $E_x = \rho_{xx}(\alpha)J_x$; for the same current density, it also induces an electric field E_y or a finite voltage along the y axis if ρ_{yx} is not zero. The latter effect is the planar Hall effect due to an in-plane magnetization and anisotropic resistivity. It has a completely different physical origin from that of the regular Hall effect which is caused by the Lorentz force from a perpendicular magnetic field.

Many of the matrix elements actually vanish due to the symmetry of the cubic crystal and the Onsager relation.¹⁷ If we apply this equation to our device geometry, where the voltages along $[110]$ and $[-110]$ are measured with a current flowing along $[110]$, then,

$$\begin{aligned} \rho_{xx} = E_{xx}/J = C_0 + 1/2C_1 + 3/8C_2 + 1/2C_4 \cos 2\phi_M \\ - 1/8C_2 \cos 4\phi_M = \rho_0(1 + A_u \cos 2\phi_M + A_c \cos 4\phi_M), \end{aligned} \quad (4)$$

$$\rho_{xy} = E_{xy}/J = 1/2(C_1 + C_2)\sin 2\phi_M = \rho_1 \sin 2\phi_M, \quad (5)$$

where C_0 , C_1 , C_2 , and C_4 are a set of constants that is related to the coefficients in Eq. (3) by

$$\begin{aligned} C_0 &= a_{11} + a_{1122} + a_{111122}, & C_1 &= a_{1111} - a_{1122} - 2a_{112211}, \\ C_2 &= a_{111111} + 2a_{111122} - a_{112211}, \\ C_4 &= a_{2323} + a_{111212}. \end{aligned}$$

These four constants can be determined from A_u , A_c , ρ_0 , and ρ_1 , which can be obtained by fitting the experimental data. As a direct consequence of the crystal symmetry, ρ_{xx} contains

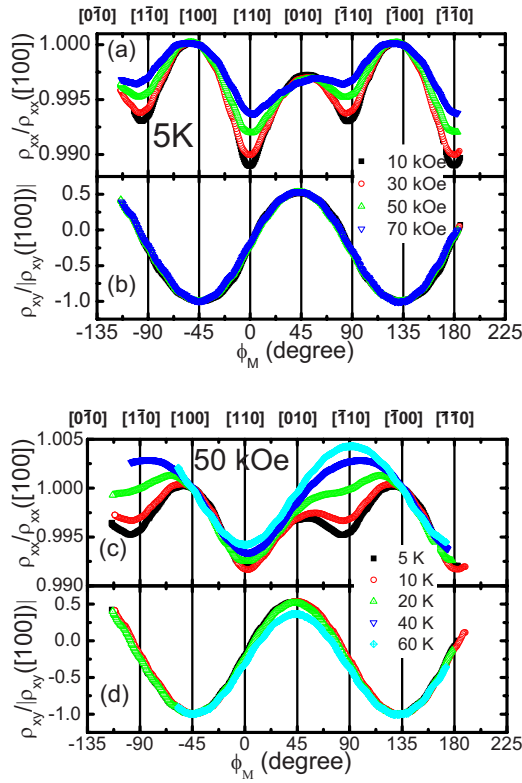


FIG. 3. (Color online) (a) ρ_{xx} and (b) ρ_{xy} at a set of fields for $T=5$ K; (c) ρ_{xx} and (d) ρ_{xy} at different temperatures for a fix field $H=50$ kOe. ρ_{xx} and ρ_{xy} are normalized to the resistivity value at $M\parallel[100]$.

both cubic and uniaxial symmetry terms, whereas ρ_{xy} contains only the uniaxial symmetry term in Eqs. (4) and (5), which agrees well with our experiments.

Now, let us examine how these two anisotropy terms depend on the magnetic field and temperature. Figures 3(a) and 3(b) show ρ_{xx} and ρ_{xy} vs ϕ_M for a series of fields $H=10, 30, 50,$ and 70 kOe measured at 5 K. The y axis shows the scaled resistivity normalized to the resistivity value when H is parallel to the $[100]$ direction. Clearly, both uniaxial and cubic anisotropies coexist at all H , but the oscillation amplitude steadily decreases at higher fields. Note that the total resistivity also decreases; therefore, the absolute resistance swing decreases even more at high fields. By decomposing the total resistivity into the cubic and uniaxial terms, we find that A_c decays much faster than A_u as H increases. In contrast, the ratio $\rho_{xy}/\rho_{xy}[100]$ does not change within experiment uncertainty since all $\rho_{xy}/\rho_{xy}[100]$ curves fall on top of each other for fields up to 70 kOe. No measurable cubic symmetry is present at these fields.

At a fixed field of 50 kOe, ρ_{xx} and ρ_{xy} are displayed for different temperatures in Figs. 3(c) and 3(d). An obvious feature is that the fourfold oscillation steadily diminishes as the temperature increases, and it disappears around T_C . This strongly suggests that the cubic symmetry originates from the long-rang FM order. In contrast, the twofold oscillation persists even above T_C where the FM order no longer exists. However, both the symmetry and amplitude of $\rho_{xy}/\rho_{xy}[100]$ remain unchanged below T_C and only show weak temperature dependence above T_C .

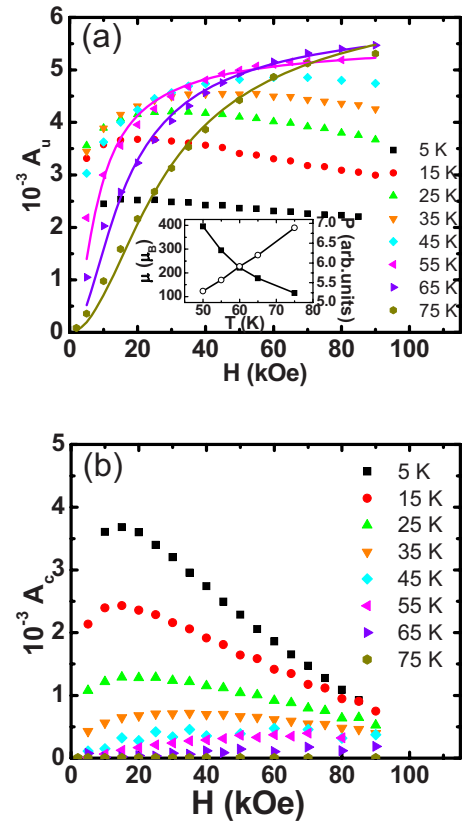


FIG. 4. (Color online) Magnetic field dependence of coefficients (a) A_u and (b) A_c for $\text{Ga}_{0.0941}\text{Mn}_{0.059}\text{As}$, obtained from fitting. The inset of (a) shows the magnetic moment of each cluster (filled square) and the contribution from all clusters, P (open circle), to A_u . The solid line in (a) is the fit for $T > T_C$, described in the text.

In nonmagnetic crystals, MR can show different behaviors when a current flows along two orthogonal crystallographic directions. In FM crystals, this MR anisotropy adds to the usual AMR which is sensitive to the direction of \mathbf{M} . This MR and its anisotropy have been attributed to the Lorentz force and follow Kohler's rule.¹⁸ The Lorentz-force induced MR or intrinsic MR is always positive. However, the MR effect observed in our experiments clearly does not share the same origin with the intrinsic MR since ours is always negative, as shown in Fig. 1(b). Moreover, it exhibits a more complex temperature dependence [Fig. 4(a)]. Although the negative MR was previously attributed to the weak localization effect,¹² it cannot easily explain why it depends on the crystal orientations and the relative direction between the current and the magnetic field.

The extracted coefficients A_u and A_c are plotted as functions of the magnetic field at various temperatures in Fig. 4 and also replotted as functions of the temperature for different fields in Fig. 5. A_c shows a monotonic and drastic decrease for all magnetic fields as T approaches T_C from below, indicating that the cubic symmetry is induced by the FM phase. A similar result has been also reported recently.¹⁹ In contrast, A_u shows nonmonotonic temperature dependence at those fields. In order to explain this complex temperature dependence of the twofold symmetry, we propose the following two-component model. We assume that at $T < T_C$, in ad-

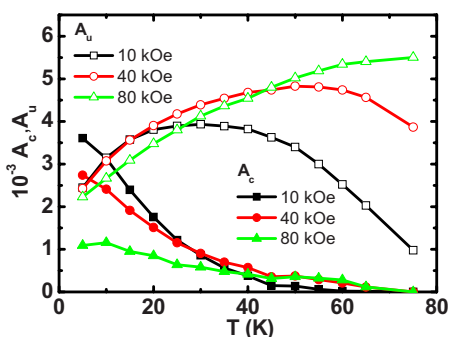


FIG. 5. (Color online) Temperature dependence of coefficients A_u (open symbols) and A_c (solid symbols) for $\text{Ga}_{0.0941}\text{Mn}_{0.059}\text{As}$ at 10, 40, and 80 kOe, obtained from fitting using Eq. (4).

dition to a long-range FM component, there exists a second component that is comprised of individual magnetic clusters. One realization of this two-component picture could be a magnetic polaron. More explicitly, since the FM coupling between Mn ions ($S=5/2$) is mediated through the delocalized holes in GaMnAs, each Mn^{2+} ion and its neighboring free holes behave like a magnetic cluster, known as a bound magnetic polaron (BMP). This carrier-mediated interaction distinguishes the ferromagnetic semiconductors from the conventional ferromagnets where the Heisenberg direct exchange links the adjacent spins. As the temperature is lowered below T_C , BMPs gradually increase in size and form the percolating network, which is the long-range FM phase. In the meantime, the BMPs isolated from the FM network behave like fragmented superparamagnetic (SPM) particles. This picture of the mixed FM and SPM is supported by Monte Carlo simulations^{20,21} and recent experiments,^{22,23} it also successfully explains the high-field resistivity behavior in InMnSb.¹¹ This two-phase model makes physical sense due to the inevitable inhomogeneous Mn distribution, which leads to the relatively hole-rich FM regions coexisting with the less hole-rich SPM regions.

Note that even in the SPM phase, there should be a non-zero twofold anisotropy term in MR. The external magnetic field can align the randomly distributed BMP moments. As the sample is rotated in H , the aligned moment sees the twofold anisotropy with respect to the electric current. It is analogous to the usual AMR and can also be described by Eq. (1). At $T > T_C$, the SPM phase is the only source of the anisotropy in ρ_{xx} and the situation is much simpler. We attempt to analyze the $T > T_C$ data since we only have to deal with the SPM component. Note that in Eq. (1), the twofold AMR due to the SPM clusters with moment μ is propor-

tional to the square of the aligned total moment, i.e., $\cos^2(\phi_M) = (M/M_s)^2$. For SPM particles, the moment M is described by the Langevin function $M/M_s = L(x) = \coth x - 1/x$, x being $\mu H/k_B T$. A decrease in T or an increase in H can enhance the magnetic moment to produce a large MR effect. We fit $A_u = P \times [L(\mu H/k_B T)]^2$ to the high-temperature field-dependence data and extract the particle moment μ and total number of particles, P , both of which are displayed in the inset of Fig. 4(a). As T increases, the individual BMP cluster size ($\sim \mu$) decreases as the carrier-mediated exchange interaction weakens; meanwhile, the total number of BMPs increases. The size of BMPs is estimated to be about 6 nm at 50 K. A somewhat larger BMP size was reported earlier for $T < T_C$.²³ Since the BMP size decreases rapidly above T_C , we believe our result is still reasonable.

For $T < T_C$, the percolating FM phase starts to contribute to ρ_{xx} , which leads to a nonzero A_c , as shown in Fig. 5. Additionally, the FM phases also contribute to the twofold anisotropy in ρ_{xx} , as seen in Eq. (4). Although both FM and SPM phases contribute to the twofold symmetry or A_u , they have opposite trends in temperature due to the expansion of the FM phase and the shrinkage of the SPM phase at low temperatures. The overall temperature dependence of twofold symmetry depends on the competition between these two phases below T_C . As a result, a peak in A_u appears at the intermediate temperatures, as shown in Fig. 5. The peak position shifts to higher temperatures as the field strength increases because the applied field enhances the contribution from the SPM phase at high temperatures. Here, we do not attempt to fit the resulting temperature dependence of A_u for lack of detailed knowledge of the two phases below T_C . A_c is suppressed by a magnetic field at all temperatures. We do not have a satisfactory explanation for this field effect.

In summary, we have found a strong magnetocrystalline anisotropic resistance effect in epitaxial GaMnAs films. The fourfold term in longitudinal magnetoresistance is related to the long-range FM order at $T < T_C$, whereas the twofold term originates from both FM and SPM spins. This effect may be used in investigating ferromagnetic ordering mechanism in other dilute magnetic semiconductors and ferromagnetic oxides.

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