Interplay between Carrier and Impurity Concentrations in Annealed $Ga_{1-x}Mn_xAs$: Intrinsic Anomalous Hall Effect

S. H. Chun,[1,2,3] Y. S. Kim,[4] H. K. Choi,[3,4] I. T. Jeong,[4] W. O. Lee,[3,4] K. S. Suh,[3,4] Y S. Oh,[3,4] K. H. Kim,[3,4] Z. G. Khim,[4] J. C. Woo,[4] and Y. D. Park[3,4,*]

[1]*Department of Physics and Institute of Fundamental Physics, Sejong University, Seoul 143-747, Korea*
[2]*Future Technology Research Division, Korea Institute of Science and Technology, Seoul 130-791, Korea*
[3]*CSCMR, Seoul National University NS 53, Seoul 151-747, Korea*
[4]*Department of Physics and Astronomy, Seoul National University NS 50, Seoul 151-747, Korea*

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Investigating the scaling behavior of annealed $Ga_{1-x}Mn_xAs$ anomalous Hall coefficients, we note a universal crossover regime where the scaling behavior changes from quadratic to linear. Furthermore, measured anomalous Hall conductivities in the quadratic regime when properly scaled by carrier concentration remain constant, spanning nearly a decade in conductivity as well as over 100 K in T_C and comparing favorably to theoretically predicated values for the intrinsic origins of the anomalous Hall effect. Both qualitative and quantitative agreements strongly point to the validity of new equations of motion including the Berry phase contributions as well as the tunability of the anomalous Hall effect.

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A rudimentary explanation of the Hall effect, attributed simply to moving charge carriers experiencing a Lorentz force, ''pressing electricity'' [1], spectacularly fails to explain even the simplest of the ferromagnetic materials. From the original measurements of iron foils by Hall [2] to complex correlated oxide systems [3,4] to graphene [5], the familiar Hall effect requires an appellation as ''ordinary'' in a sea of extraordinary effects, colorfully termed as ''anomalous'' to ''quantum,'' and even ''extraordinary.'' Only recently has the subtle role of the quantum geometry of the Fermi surface been recognized as intrinsic origins for much of these effects [6,7]. As any material property determined by transport measurements, the Hall effect reflects contributions from both intrinsic and extrinsic mechanisms. The separation of the two and the microscopic origins responsible for each have been a source of great contention for decades, especially in magnetic materials, and nonreduced dimensioned materials in general, with limited experimental observations [8] of a clear intrinsic mechanism for the anomalous Hall effect (AHE).

In the area of semiconductor spintronics, AHE has had an important role in both demonstrating the novelty of carrier mediated ferromagnetic ordering, an interplay between carrier concentration and magnetic properties, in diluted magnetic semiconductors (DMS) [9] and indirect characterization of magnetic properties [10,11]. Furthermore, a special case of the AHE with vanishing spin polarization has recently received much interest as possible sources of spins in a spintronic device [12]. The idea of dissipationless intrinsic Hall current can be traced to Karplus-Luttinger (KL) formalism [13] in which the term ''anomalous velocity'' has been recently reinterpreted as a manifestation of the Berry curvature of occupied electronic Bloch states [14]. Following KL arguments, the anomalous Hall coefficient (R_S), related to the strength of the spin-orbit coupling, scales quadratically with longitudinal resistivity (ρ_{xx}), similar behavior to the extrinsic side-jump mechanism in which the carrier is asymmetrically displaced by impurity scattering, proposed by Berger [15]. In between, Smit argued that R_S must vanish in a periodic lattice and proposed the extrinsic skew-scattering mechanism, which predicts the transverse resistivity (ρ_{xy}) to scale linearly with ρ_{xx} [16].

Here, we report a clear and distinct crossover in the underlying mechanisms for the AHE in a series of annealed $Ga_{1-x}Mn_xAs$, as evidenced by the changing scaling behavior of the anomalous Hall coefficient. In the regime where the scaling behavior is quadratic, we observe the transverse conductivity (σ_{xy}) when properly scaled to be independent of longitudinal resistivity (ρ_{xx}), and that the measured values compare well to recently proposed theories on the intrinsic origins of AHE evoking the Berry phase [7]. Furthermore, the anomalous Hall current, impervious to ρ_{xx}, can be manipulated by implicitly and explicitly varying the carrier concentration in $Ga_{1-x}Mn_xAs$.

$Ga_{1-x}Mn_xAs$, a ferromagnetic semiconductor, is one of the most intensively studied materials in the context of semiconductor spintronics, and the recipe for its growth is well known [10,17,18]. For our study of the AHE in $Ga_{1-x}Mn_xAs$ [19,20], we explicitly vary the total Mn concentration (N_{Mn}) between 2.4% and 6.1% during low-temperature molecular beam epitaxial growth (LT-MBE). Furthermore, we implicitly vary other impurity concentrations by low-temperature annealing with temperatures ranging from 200–350 °C. After growth, the samples are fashioned into electrically isolated 300 μm $\times$ 1900 μm Hall bar structures. Samples are then annealed in a tube furnace in a flowing dry N_2 environment for 1 h with annealing temperature, measured by a thermocouple near the sample. After annealing, indium contacts are fashioned

and verified as Ohmic for transport measurements in a closed-cycle cryostat and/or in a quantum design physical property measurement system (equipped with a 7 T magnet) with customized ac lock-in technique capabilities (excitation current of 10–40 μA from a current calibrator at 17 Hz) [Fig. 1(a)].

Similar to reports by others [21], we observe the magnetic properties, in terms of the highest magnetic ordering temperatures (T_C), and transport properties, in terms of lowest resistivities, to be optimized at an annealing temperature of $\sim$250 °C with further deterioration of such properties at higher annealing temperatures [Fig. 1(b)]. Both our transport and magnetic measurements are consistent with a widely held view that low-temperature annealing initially removes Mn_I (N_{Mn}^{++}) and other electrically active donor impurities as evidenced by increases in longitudinal conductivity (σ_{xx}), hole carrier concentration (n_h), and T_C. Annealing above optimal temperatures results in deterioration of such properties as $\mathrm{Mn}_{\mathrm{Ga}}(N_{\mathrm{Mn}}^{-})$, the source of both spins and carriers, decreases. Even for the highest annealing temperature considered (350 °C), magnetization data along with high resolution x-ray diffraction θ–2θ measurements do not show secondary ferromagnetic phases. However, the existence of such phases cannot be fully ruled out [20]. In short, after LT-MBE and low-temperature annealing, a series of $\mathrm{Ga}_{1-x}\mathrm{Mn}_x\mathrm{As}$ samples exhibiting both insulator- and metallic-like behaviors with σ_{xx} ranging more than 2 orders of magnitude and T_C ranging from $\sim$25 K to $\sim$160 K was obtained.

A qualitative means to ascertain the origins of the AHE is to plot the scaling relationship of either ρ_{xy} or R_S to ρ_{xx}. The scaling relationship ($\rho_{xy} = c\rho_{xx}^n$) as applied to $\mathrm{Ga}_{1-x}\mathrm{Mn}_x\mathrm{As}$ is problematic, as magnetization is inevitably related to ρ_{xx} by n_h as well as weak localization of carriers for higher resistive samples [22]; thus, the relationship $R_S = c\rho_{xx}^n$ may be more appropriate. In AHE literature, the scaling relationship of either ρ_{xy} or R_S to ρ_{xx} is plotted by varying ρ_{xx} by measurement temperature [i.e., $\rho_{xx}(T)$], by magnetic solute concentration [i.e., $\rho_{xx}(N_{\mathrm{Mn}}^{-})$], or by both. The general relationship between R_S and ρ_{xy} is given from the empirical relationship as a sum of the ordinary and anomalous components:

$$\rho_{xy} = R_o B_Z + \mu_o R_S M_Z, \tag{1}$$

where R_o is the ordinary Hall coefficient from which carrier concentration and type can be discerned ($\sim 1/qn_h$) and M_Z is the magnetization of the sample along the standard Hall geometry where the magnetic field is applied perpendicular to the plane. This empirical relationship is upheld for both intrinsic and extrinsic origins of the AHE [1,8,14].

Measurement of ρ_{xy} allows for an indirect means to characterize magnetic properties, especially for $\mathrm{Ga}_{1-x}\mathrm{Mn}_x\mathrm{As}$ with low resistivities [Figs. 1(c) and 1(d)]. From the empirical relationship [(Eq. (1)], values of R_S in principle can be determined from the y intercept ($\sim \mu_o R_S M_S$) of the Hall response [Fig. 2(a)] with magnetic saturation values (M_S) measured separately. In practicality, due to intrinsic magnetoresistances [Fig. 2(b)] and difficulties in achieving technical magnetization saturation, especially for insulating samples, while evaluating R_S of metallic samples are nearly error free, that of insulating samples suffers from overestimation by extrapolating the high field response of ρ_{xy}. Even thus, plotting $\log(R_S)$ vs $\log(\rho_{xx})$ at 15 K [Fig. 2(c)] shows a clear demarcation where a fit to n in the scaling relationship ($R_S = c\rho_{xx}^n$) changes from a value of 2.05 to 1.30, for $\rho_{xx} > \sim$10 mΩ cm as similarly alluded by Ruzmetov *et al.*

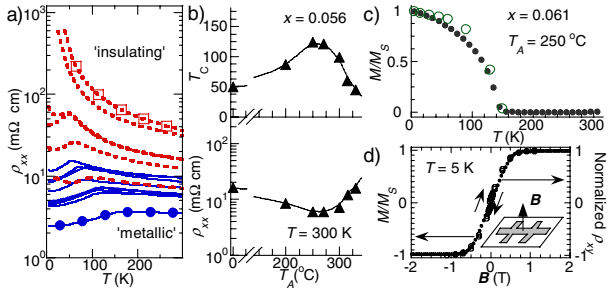

FIG. 1 (color online). (a) $\mathrm{Ga}_{1-x}\mathrm{Mn}_x\mathrm{As}$ samples that exhibit ''metalliclike'' behavior [circle (blue) solid] and that exhibit ''insulatinglike'' behavior [square (red) dashed] are identified by the sign of $\partial\rho_{xx}/\partial T$ far below T_C. (b) For $x = 0.056$ annealed for 1 h, T_C (upper) and ρ_{xx} (lower) are plotted for various annealing temperatures. (c) For $x = 0.061$ annealed at 250 °C, temperature dependence of normalized magnetization from SQUID measurements (solid circle) and from Arrott plots of AHE data (open circle) are plotted. (d) For as-grown $x = 0.061$, field dependence from SQUID magnetometry measurements (open circle) with applied magnetic fields normal to the epifilm which reflects AHE (solid circle).

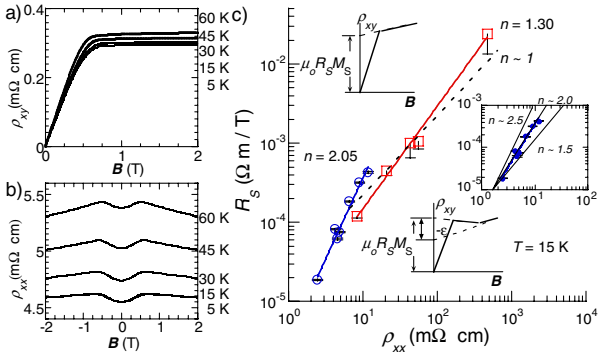

FIG. 2 (color online). (a),(b) For as-grown $x = 0.061$, ρ_{xy} (a) and ρ_{xx} (b) dependences to applied magnetic field below T_C. (c) For a measurement temperature of 15 K, R_S dependence to ρ_{xx} is plotted for all samples with differing $N_{\mathrm{Mn}}^{-}/N_{\mathrm{Mn}}^{++}$. Line representing $n = \sim 1$ is provided as a guide. Insets depict methods used to determine R_S, and the major errors associated, for metallic (top) and insulating (bottom) samples as well as quadratic scaling behavior with $n = \sim$1.5, $\sim$2, $\sim$2.5 lines as a guide.

[23]. Although some groups have inferred intrinsic origins of AHE in $Ga_{1-x}Mn_xAs$ by observing $n \sim 2$ to the scaling relationship $\rho_{xy} = c\rho_{xx}(T)^n$ by varying the measurement temperature for a fixed N_{Mn}^- and resulting in marginal variation of ρ_{xx} [24], here we vary the magnetic solute concentrations (N_{Mn}^-) to vary ρ_{xx} over 2 orders of magnitude at a fixed low temperature to minimize localization effects, especially for insulatorlike samples. But, whether the anomalous Hall effect is intrinsic for $Ga_{1-x}Mn_xAs$ is not clear by examining the scaling relationship alone [25].

Theoretical treatments on the origins of the AHE predict various scaling relationships. The observed quadratic relationship between R_S and ρ_{xx} for $\rho_{xx} < \sim 10$ mΩ cm can be interpreted as a manifestation of either intrinsic or extrinsic origins such as the side-jump mechanism. Approximately linear relationship for $\rho_{xx} < \sim 10$ mΩ cm can be only attributable to dominance of extrinsic origins, either from skew-scattering mechanism or from phonon-assisted hopping of holes between localized states [26]. It is also worth noting that the length scales of the extrinsic side-jump mechanism (0.01–0.1 nm) [15] is thought to be orders of magnitude smaller than skew scattering with predominance of transition from linear to quadratic scaling behavior for increasing resistivities for magnetic systems dominated by extrinsic origins [1]. As the observed quadratic scaling relationship attributable to intrinsic origins for $Ga_{1-x}Mn_xAs$ occurs for resistivities much larger than metallic samples such as $SrRuO_3$ [4], we require further analysis to discern our observed scaling relationships as from extrinsic or intrinsic origins. Further comparisons are made possible by a recent theory by Jungwirth, Niu, and MacDonald (JNM) [7] evoking the Berry phase in the momentum space.

In brief, JNM calculated σ_{xy} exactly with the assumptions of infinite spin-orbit coupling strength and a mean-field Hamiltonian with the effective field h is given by$N_{Mn}SJ_{pd}$, where N_{Mn} is the density of Mn ions with spin $S = 5/2$:

$$CN_{Mn}J_{pd}n_h^{-1/3} < \sigma_{xy} < 2^{2/3}CN_{Mn}J_{pd}n_h^{-1/3}, \qquad (2)$$

where $C = \frac{5}{2}\frac{e^2}{2\pi\hbar}\frac{(3\pi^2)^{-1/3}}{2\pi\hbar^2}m_{hh}$. The upper and lower bounds of σ_{xy} correspond to $m_{lh} \approx m_{hh}$ and $m_{lh} \ll m_{hh}$, respectively, with σ_{xy} of $Ga_{1-x}Mn_xAs$ being closer to the lower bound ($m_{lh}/m_{hh} = 0.16$). In JNM calculations, there exists a tacit condition for a "clean limit" where all of N_{Mn} participate in the magnetic ordering as well as later inclusions of finite spin-orbit coupling, warping of band structures; and strains and defects in applying to experimental data [27].

One difficulty in an accurate experimental verification of the above relationship is an accurate determination of n_h, another being determination of N_{Mn} and a clean-limit condition, which we will address later. In JNM's work, their theoretical calculation of σ_{xy} was compared to one sample from which n_p, and consequently N_{Mn}^- as some fraction of N_{Mn}, was measured at 50 mK with an applied magnetic field of 27 T [18]. In addition to similar practical problems for an accurate determination of R_S, determination of n_h further suffers from the influence of the anomalous term even for temperatures much greater than T_C. Recently, Ruzmetov *et al.* cleverly reexpressed the empirical relationship [Eq. (1)] for $T > T_C$, with paramagnetic behavior expressed in terms of the Curie-Weiss Law and found their model estimation of n_h to be comparable to values obtained from high fields (above 25 T) and electrochemical capacitance measurements [23]. Then, we follow methods outlined by Ruzmetov *et al.* with our fitting parameter n [28].

In our scheme to vary ρ_{xx}, we explicitly varied N_{Mn} during LT-MBE growth and implicitly varied N_{Mn}^{++} and N_{Mn}^- by low-temperature annealing. In the course of determining simple scaling relationships of R_S to ρ_{xx}, we observe much of the transport properties and magnetic properties to have a distinct dependence on n_h [Figs. 3(a) and 3(b)], as expected from a carrier mediated DMS as $Ga_{1-x}Mn_xAs$ [10]. Moriya and Munekata found that N_{Mn}^- becomes saturated despite the steady increase of N_{Mn}, and consistent scattering coefficients when N_{Mn}^- was used in the room-temperature AHE analysis [29]. Magnetization study also supports this notion: the seemingly deficient magnetization is recovered if only the ionized Mn atoms are counted [30].

Therefore, it is concluded that N_{Mn}^- or n_h rather than N_{Mn} characterizes $Ga_{1-x}Mn_xAs$ epilayers. The monotonic dependences of T_C and ρ_{xx} on n_h inferred further support the argument. Then, we find that [Eq. (2)] can be adapted to explain the behavior of metallic $Ga_{1-x}Mn_xAs$ samples once we replace N_{Mn} with n_h. Equation (2) then simplifies to: $\sigma_{xy} = CJ_{pd}n_h^{2/3}$ if we take the lower bound. Thus, we normalize σ_{xy} by $n_h^{2/3}$ and the results are shown [Fig. 4(a)]. Clearly the two classes (metallic and insulating) of $Ga_{1-x}Mn_xAs$ express differing behaviors with the same crossover as the change in scaling behavior of R_S. While the insulating samples show drastic changes as T_C varies,

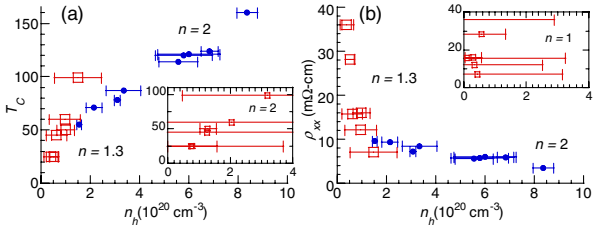

FIG. 3 (color online). (a),(b) Dependence of T_C (a) and ρ_{xx} (at 300 K) (b) to n_h is plotted for samples with differing N_{Mn}^-/N_{Mn}^{++}. The carrier concentration is found as outlined in Ref. [23]. Insets depict T_C or ρ_{xx} dependence to n_h fitted with $n = 2$ (a) and $n = 1$ (b) with great uncertainty from fitting compared to $n = 1.3$ for insulating samples.

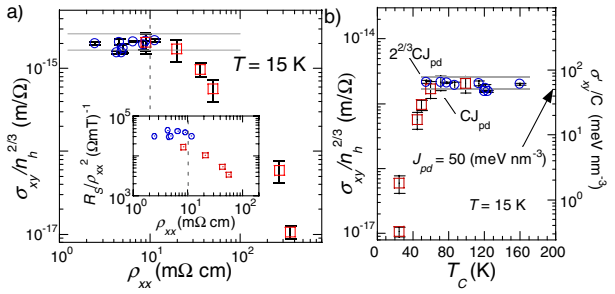

FIG. 4 (color online). (a),(b) Normalized σ_{xy} (measured at $T = 15$ K) to $n_h^{2/3}$ as function of ρ_{xx} (a) and T_C (b) is plotted. Inset in (a) depicts $\frac{R_S}{\rho_{xx}^2} \approx \frac{\sigma_{xy}}{\mu_o M_S}$. Scaling behaviors for metallic and insulating samples are drastically different along with values of $\sigma_{xy}/n_h^{2/3}$ for metallic samples comparing favorably to JNM calculations (b).

the metallic samples show a good scaling behavior despite the large change in T_C from 50 to 160 K [Fig. 4(b)]. The most striking observation is the reasonable quantitative agreement with CJ_{pd}, when we use the widely accepted value of $J_{\mathrm{pd}} = 50$ meV nm^{-3}.

To summarize, our data clearly show a universal crossover between the scaling behaviors of the anomalous Hall coefficient. The linear scaling behavior can be attributed to dominance of extrinsic origins. In the quadratic regime, we find the Hall conductivities to be independent of impurity concentrations and to agree favorably, qualitatively, and quantitatively, with JNM calculations in the clean limit for intrinsic origins of the AHE in $Ga_{1-x}Mn_xAs$. Our results signify another importance on the role of carrier and impurity concentrations in $Ga_{1-x}Mn_xAs$ diluted magnetic semiconductors. Carriers and impurities govern the differing origins of the anomalous Hall effect.

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*Electronic address: parkyd@phya.snu.ac.kr

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[28] The phenomological expression in Ref. [23] uses scaling parameter $n = 2$ to fit n_h for samples with $\rho_{xx} <$ 10 mΩ cm for $T > T_C$, in which temperature regime, we feel that $n \sim 1$ has more physical meaning. For $T > T_C$, $Ga_{1-x}Mn_xAs$ would be more akin to a paramagnetic matrix embedded with magnetic impurities [A. Fert and O. Jaoul, Phys. Rev. Lett. **28**, 303 (1972)]. In our fits for metallic samples, we see marginal differences between values of n_h from fitting with n as well as from R_o. However, as depicted in Fig. 3 insets, for insulating samples, there is a large variance dependent on the chosen n.
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