Interplay between carrier and impurity concentrations in annealed Ga$_{1-x}$Mn$_x$As intrinsic anomalous Hall Effect

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Investigating the scaling behavior of annealed Ga$_{1-x}$Mn$_x$As anomalous Hall coefficients, we note a universal crossover regime where the scaling behavior changes from quadratic to linear, attributed to the anomalous Hall Effect intrinsic and extrinsic origins, respectively. Furthermore, measured anomalous Hall conductivities when properly scaled by carrier concentration remain constant, equal to theoretically predicated values, spanning nearly a decade in conductivity as well as over 100 K in $T_C$. Both the qualitative and quantitative agreement confirms the validity of new equations of motion including the Berry phase contributions as well as tunability of the intrinsic anomalous Hall Effect.

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Rudimentary explanation of the Hall Effect, attributed simply to moving charge carriers experiencing a Lorentz force, ‘pressing electricity’ $^\text{1}$, spectacularly fails to explain even the simplest of the ferromagnetic materials. From the original measurements of iron foils by E.H. Hall$^2$ to complex correlated oxide systems$^3$ to graphene$^4$, the familiar Hall Effect, from which the nature and amount of carriers can be determined, 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$^5,6$. As any material property determined by transport measurements, the Hall Effect reflects contributions from both intrinsic and extrinsic mechanisms. Separation of the two and the microscopic origins responsible for each has been a source of great contention for decades, especially in magnetic materials, and non-reduced dimensioned materials in general, with limited experimental observations$^2,5$ 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 in diluted magnetic semiconductors (DMS)$^6$ and indirect characterization of magnetic properties$^{10,11}$. Furthermore, a special case of the AHE with vanishing spin polarization, the intrinsic spin Hall Effect$^{12}$, has recently received much interest as possible sources of spins in a spintronic device, utilizing the dissipationless nature of the intrinsic transverse spin current$^{13}$. The idea of dissipationless intrinsic Hall current can be traced to Karplus-Luttinger (KL) formalism$^{14}$ in which the term ‘anomalous velocity’ has been recently reinterpreted as a manifestation of the Berry curvature of occupied electronic Bloch states. Following KL arguments, the anomalous Hall coefficient ($R_S$), related to the strength of the spin orbit coupling, scales quadratically with $\rho_{xx}$, 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 ($\rho_{xy}$) to scale linearly with $\rho_{xx}$,$^{16}$. As much experimental evidence showed both linear scaling relationships as well as quadratic, in higher resistive samples, AHE for decades had been thought as an extrinsic phenomenon related to impurity scattering, and KL ideas of intrinsic origins had been discounted. With recent evolution of the Berry phase in the momentum space as the intrinsic origins of AHE, in addition to predicted scaling relationships, measured $\sigma_{xy}$ can be directly compared to values from well-known material parameters and fundamental quantities to discern intrinsic dissipationless spin currents.

Here, we report a clear and distinct cross-over in the underlying mechanisms for the AHE in a series of annealed Ga$_{1-x}$Mn$_x$As. In the intrinsic regime, we observe the transverse conductivity ($\sigma_{xy}$) when properly scaled to be independent of longitudinal resistivity ($\rho_{xx}$), and that the measured values fit recently proposed theories on the intrinsic origins of AHE evoking the Berry phase$^6$. Furthermore, the intrinsic dissipationless spin Hall current, impervious to $\rho_{xx}$, can be manipulated by implicitly and explicitly varying the carrier concentration in Ga$_{1-x}$Mn$_x$As.

Ga$_{1-x}$Mn$_x$As, a ferromagnetic semiconductor, is one of the most intensively studied materials in the context of semiconductor spintronics, and the recipe for
FIG. 1: (a&b) Series of Ga$_{1-x}$Mn$_x$As samples with varying $N_{Mn}$ and $N_{Mn}$ that exhibit ‘metallic’-like behavior ((a) circle) and that exhibit ‘insulating’-like behavior ((b) square) are identified by the sign of $\partial \rho_{xx}/\partial T$ far below $T_C$. (c) Plot of magnetic ($T_C$) and transport ($\sigma_{xx}$, open square) properties as function of carrier concentration ($n_h$). 

Plotting all the measured $\rho_{xx}$ and $\rho_{xy}$ (Fig. 2), the data indicate an existence of an overall scaling relationship akin to the skew scattering origins for the AHE even though $\rho_{xx}$ reflects a wide variance of impurity concentrations. The scaling relationship ($\rho_{xy} = c \rho_{xx}^m$) as applied to Ga$_{1-x}$Mn$_x$As is problematic, as magnetization is inevitably related to $\rho_{xx}$ by $n_h$ as well as weak localization of carriers for higher resistive samples [23]; thus, the relationship $R_S = c \rho_{xy}^p$ may be more appropriate. It has been widely reported that the magnetization of Ga$_{1-x}$Mn$_x$As, even for metallic samples, does not easily saturate, thus making a differentiation between the ordinary term and the anomalous term difficult. Thus, we estimate $n_h$ by fitting the temperature dependence of $\rho_{xy}$ [24]. With the anisotropy of the Ga$_{1-x}$Mn$_x$As epilayer in relation to the applied magnetic field and with $R_S$ much larger than the ordinary Hall coefficient ($R_o$), we determine $R_S$ as $(\partial \rho_{xy}/\partial B)(\mu_o \partial M_Z/\partial B_Z)^{-1}$ from $\rho_{xy} = R_o B + \mu_o R_S M$, an empirical relationship which is upheld for both intrinsic and extrinsic origins of AHE. For the range of $x$ considered in this study, we expect $\partial M_Z/\partial B_Z$ to be constant $(\sim 1/\mu_o)$ from anisotropic arguments along the lines of Liu et al. and Titova et al. [27]. These two studies find the perpendicular uniaxial anisotropy fields to be nearly independent of $n_h$ and the cubic anisotropy terms to be negligible due to the large built-in strain during LT-MBE. Thus, we estimate $c R_S$ as $\partial \rho_{xy}/\partial B$ (Fig. 2b).

Plotting $\log(c R_S)$ vs. $\log(\rho_{xx})$ (Fig. 4a) shows a clear demarcation where a fit to $n$ in the scaling relationship ($R_S = c \rho_{xx}^p$) changes from a value of two to one, for $\rho_{xx} > 10$ m$\Omega$-cm as similarly alluded by Ruzmetov et al. [24], especially at lower temperatures (Fig. 4b). Here, we vary both temperature and magnetic solute concentrations ($N_{Mn}$) to vary $\rho_{xx}$. Furthermore, for temperatures near $T_C$ (Fig. 4b inset), the scaling relationship seems to
align the magnetic fields below respective \( T_C \)'s are plotted along with data from two other groups (ref. 17 & 21). Each cluster of data points represents an isotherm of a particular annealed Ga\(_{1-x}\)Mn\(_x\)As (inset for S6.1-250) \((n \approx 1 \text{ line as a guide}).\)

converge to a value of \( n \) equal to one, a result analogous to skew scattering origins of AHE in paramagnetic matrix with embedded magnetic impurities, or possibly due to phonon-assisted hopping of holes between localized states in the impurity band. For some of the highest annealing temperatures, we note \( n > 3 \), suggestive of inhomogeneous systems. Whether the dissipationless anomalous Hall current is intrinsic for all Ga\(_{1-x}\)Mn\(_x\)As samples is not clear by examining the scaling relationships alone. Lee et al. report of the dissipationless intrinsic AHE in the ferromagnetic spinel CuCr\(_2\)Sc\(_{4-x}\)Br\(_x\) maintains that a quadratic relationship between \( R_S/n_h \) and \( c\rho_{xx} \) should be expected. Interestingly, a similar plot (Fig. 3a) indicates quadratic relationship between \( R_S/n_h \) and \( \rho_{xx} \) for all samples considered except for two samples annealed at the highest temperature, \( 350^\circ \text{C} \).

The unexpected quadratic relationship between \( R_S/n_h \) and \( \rho_{xx} \) for Ga\(_{1-x}\)Mn\(_x\)As questioned us what should be the proper normalization method for Ga\(_{1-x}\)Mn\(_x\)As. The answer comes from the intrinsic AHE theory developed by Jungwirth, Niu, and MacDonald (JNM). In brief, they use a semi-classical transport theory including the effect of Berry phase:

\[
\dot{x} = \frac{1}{\hbar} \frac{\partial E_n(\vec{k})}{\partial \vec{k}} + \vec{k} \times \vec{B}_n(\vec{k}) \tag{1}
\]

\[
\hbar \dot{\vec{k}} = -e(\vec{E} + \dot{x} \times \vec{B}(\vec{x})) \tag{2}
\]

The resemblance between (Eq. 1) and (Eq. 2) is why the Berry curvature \( \vec{B}_n(\vec{k}) \) is called “the magnetic field in momentum space”. JNM calculated \( \sigma_{xy} \) \( \text{exactly with the assumptions of infinite spin-orbit coupling strength} \) and a mean-field Hamiltonian with the effective field \( \hbar \) is given by \( N_{Mn} S J_{pd} \), where \( N_{Mn} \) is the density of Mn ions with spin \( S = 5/2 \):

\[
CN_{Mn} J_{pd} n_p^{-1/3} < \sigma_{xy} < 2^{2/3} CN_{Mn} J_{pd} n_p^{-1/3} \tag{3}
\]

where \( C = \frac{2}{\hbar^2} \frac{3}{2} \frac{(3\pi^2)^{-1/3}}{m_{hh}^{1/3}} \). The upper and lower bounds of \( \sigma_{xy} \) correspond to \( m_{hh} \approx m_{hh} \) and \( m_{hh} \ll m_{hh} \), respectively, with \( \sigma_{xy} \) of Ga\(_{1-x}\)Mn\(_x\)As being closer to the lower bound \((m_{hh}/m_{hh} = 0.16)\). 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.

Again, in our scheme to vary \( \rho_{xx} \), we explicitly varied \( N_{Mn} \) during LT-MBE growth and implicitly varied \( N_{Mn}^{++} \) and \( N_{Mn}^- \) by low temperature annealing. In course of determining simple scaling relationships of \( R_S \) to \( \rho_{xx} \), we observe much of the transport properties and magnetic properties to have a distinct dependence on \( n_h \) (Fig. 3c), as expected from a carrier mediated DMS as Ga\(_{1-x}\)Mn\(_x\)As. 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. Magnetization study also supports this notion: the seemingly deficient magnetization is recovered if only the ionized Mn atoms are counted.
Therefore, it is concluded that $N_{MN}$ or $n_h$ rather than $N_{MN}$ characterizes Ga$_{1-x}$Mn$_x$As epilayers. The monotonic dependences of $T_C$ and $\sigma_{xy}$ on $n_h$ inferred further support the argument. Then, we find that (Eq. 3) is enough to explain the behavior of metallic Ga$_{1-x}$Mn$_x$As samples once we replace $N_{MN}$ with $n_h$. Equation 3 then simplifies to: $\sigma_{xy} = CJ_{pd}n_h^{2/3}$ if we take the lower bound.

Thus, we normalize $\sigma_{xy}$ by $n_h^{2/3}$ instead of $n_h$ and the results are shown (Figs. 5b&c). Clearly the two classes (metallic and insulating) of Ga$_{1-x}$Mn$_x$As express different behaviors with the same crossover as the change in scaling behavior of $R_S$.

To summarize, our data clearly shows a universal crossover between intrinsic and extrinsic in the scaling behavior of the anomalous Hall coefficient and that anomalous Hall conductivities of metallic Ga$_{1-x}$Mn$_x$As follows the theoretical prediction qualitatively and quantitatively. And, we find the intrinsic nature of AHE, akin to dissipationless spin Hall currents, in Ga$_{1-x}$Mn$_x$As to be robust.

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