## Critical behavior from the anomalous Hall effect in (GaMn)As

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An unconventional approach to scaling based on the anomalous Hall effect is presented and utilized to demonstrate conclusively that epitaxial (metallic)  $Ga_{0.98}Mn_{0.02}As$  is a mean-field ferromagnet. Such a result provides strong support for the *assertion* that the underlying interactions can be treated within the framework of Landau mean-field theory, *direct* support for which had previously been lacking. This scaling approach also provides confirmation—albeit indirect—that the origin of the anomalous Hall effect in this system is intrinsic at the composition studied.

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Magnetic semiconductors are regarded currently as an important class of materials due, for example, to their potential application as injectors for spintronic devices.<sup>1</sup> In this context, GaAs has been the object of considerable recent interest since the discovery that it becomes ferromagnetic when Ga is replaced by Mn.<sup>2-7</sup> The study of critical behavior in ferromagnets such as (GaMn)As plays a pivotal role in understanding the physical basis of such behavior.<sup>8</sup> Establishing the universality class of any material provides insight into the range of the underlying interactions,<sup>9,10</sup> from which the dominant interaction can often be inferred, for example, long/infinite range interaction in the case of mean-field exponents, as opposed to short range/near neighbor in the case of Heisenberg model values. The present study demonstrates conclusively that an epitaxial (metallic) Ga<sub>0.98</sub>Mn<sub>0.02</sub>As microstructure is a mean-field ferromagnet by utilizing an unconventional scaling approach based on the anomalous Hall effect (AHE).

While the origin of the AHE in ferromagnets is more complicated than in "normal" conductors due to additional contributions from the presence of magnetic moments,<sup>11</sup> it has been widely adopted to investigate magnetic materials.<sup>12,13</sup> Specifically, the evolution of AHE with temperature and field has been linked directly with that of the magnetization M through the well established, though empirical relationship,<sup>11–13</sup>

$$\rho_{xy} = R_0 B + 4\pi R_S M. \tag{1}$$

Here  $\rho_{xy}$  is the conventional Hall resistivity,  $B = [H + 4\pi(1 - N)M]$  while *H* is the applied magnetic field; with the latter oriented perpendicular to the current flow in samples with the present geometry, the corresponding demagnetization factor *N* typically approaches unity ( $\approx 1$ ). The "ordinary" Hall coefficient,  $R_0 = \frac{1}{nec}$  (*e* being the electronic charge), arising from carrier orbit curvature/Lorentz force effects, yields information on the carrier type and concentration (*n*).  $R_S$  is the anomalous Hall coefficient, on which this study focuses; its dependence on longitudinal resistivity,  $\rho_{xx}$ , plays a central role<sup>11-14</sup> in the analysis presented below.

As  $\rho_{xy} \ll \rho_{xx}$  in the present system, the Hall conductivity  $\sigma_{xy}$  can be written as  $\sigma_{xy} = \frac{\rho_{xy}}{\rho_{xy}^2 + \rho_{xx}^2} \approx \frac{\rho_{xy}}{\rho_{xx}^2} = \frac{R_0 H + 4 \pi R_S M}{\rho_{xx}^2}$ .<sup>13</sup> With  $\sigma_{xy}$ 

subdivided as  $\sigma_{xy} = \sigma_{xy}^O + \sigma_{xy}^A$  in the usual notation for ferromagnets,<sup>14</sup> the following expression for the anomalous Hall conductivity,  $\sigma_{xy}^A$ , results

$$\sigma_{xy}^{A} = \sigma_{xy} - \sigma_{xy}^{O} = \frac{4\pi R_{S}M}{\rho_{xx}^{2}}.$$
 (2)

As is clear from this equation, an accurate determination of the anomalous component  $\sigma_{xy}^A$ —and hence an evaluation of its dependence on the magnetization *M*—relies on a careful subtraction of the ordinary component,  $\sigma_{xy}^{O} = \frac{R_0 H}{\rho_{xx}^2}$ , an issue discussed below. Two additional-and important-points also emerge for this equation. First, once the system's magnetization is measured, it provides a means of identifying the dependence of  $R_S$  on  $\rho_{xx}$  and hence characterizing the dominant mechanism underlying the AHE in this system<sup>15</sup> [as demonstrated recently for the magnetic semiconductor  $Fe_{0.8}Co_{0.2}Si$  (Ref. 16)]. Second, essentially the converse of the above, if the dependence of  $R_s$  on  $\rho_{xx}$  can be or has been established by other means, then it is possible to determine the behavior of the magnetization M from measurements of  $\sigma_{xy}^{A}$ . The latter has important consequences in the present context, viz., in systems with reduced dimensions/ dimensionality, for which the associated magnetic signal is weak. Such an approach is implemented below, enabling the (magnetic) universality class of epitaxial  $Ga_{0.98}Mn_{0.02}As$  to be established and compared with theoretical predictions.

The occurrence of the AHE in ferromagnetic (III, Mn)V semiconductors has not only been investigated extensively<sup>1,3,4,15,17–19</sup> but theories for it in the metallic regime have also been proposed recently based on a mean-field prescription. In particular, the latter concludes the presence of an impurity scattering-independent *intrinsic* AHE in this regime, <sup>15</sup> which yields  $R_S \propto \rho_{xx}^{2}$ .<sup>17,18</sup> This dependence is confirmed indirectly by the analysis presented below; subsequently scaling behavior based on the anomalous Hall conductivity demonstrates unequivocally the applicability of a mean-field approach. Specifically, inserting the predicted dependence of  $R_S$  into Eq. (2) leads to the result that the magnetization is *directly* proportional to the anomalous Hall conductivity,  $\sigma_{xy}^A \propto M$ , enabling the field and temperature variation in M to be extracted form magnetotransport data.



FIG. 1. (Color online) Field-dependent magnetotransport measurements. (a) Hall resistivity  $\rho_{xy}$  and (b) longitudinal resistivity  $\rho_{xx}$ measured at various fixed temperatures (46–78 K in 2 K steps) in the fields up to 70 kOe. Inset is the corresponding zero-field temperature dependent  $\rho_{xx}$ . (c) The corresponding anomalous Hall conductivity  $\sigma_{xy}^A$  derived from  $\rho_{xy}$  and  $\rho_{xx}$  as discussed in the text.

Epitaxial (GaMn)As specimens were grown on a GaAs substrate, then etched into a microstructural form approximately 50 nm thick, 250 µm wide, and 1000 µm long. Previous results reporting the broadband electrical detection of spin excitations in a coincidentally prepared specimen using a photovoltaic technique have been reported elsewhere.<sup>20</sup> Magnetotransport measurements were carried out in a commercial Quantum Design physical properties measurement system with an excitation current 10  $\mu$ A at 499 Hz. The Hall bar mismatch was canceled by the field-scanning technique. A summary of these magnetotransport data in the critical regime is presented in Figs. 1(a) and 1(b). The variation in  $\rho_{xy}$  with temperature and field is strongly suggestive of a link with the magnetization, and indeed, far above the ordering temperature (and hence not reproduced here), it exhibits the same linear dependence on field as is well established for M in a wide range of system which also exhibit magnetic ordering. The temperature dependent of  $\rho_{xx}$  [inset, Fig. 1(b)] confirms that the present system lies in the metallic region,

thus validating the following discussion. The first issue that needs to be addressed in evaluating the anomalous Hall conductivity  $\sigma_{xy}^A$  as accurately as possible is the careful evaluation of the contribution arising from the ordinary Hall conductivity  $\sigma_{xy}^O = \frac{R_0 H}{\rho_{xx}^2}$ . This was done using a previously advocated technique,<sup>11</sup> i.e., the intercept of the slope of  $\rho_{xy}$  vs H,  $d\rho_{xy}/dH=R_0$  extrapolated from high fields (50 kOe < H < 70 kOe) where the susceptibility is expected to vanish. This yields a weakly temperature-dependent hole density  $n \approx (5.0 \pm 0.3) \times 10^{20}$  cm<sup>-3</sup>, consistent with previous estimates<sup>1,7</sup> while also indicating that the anomalous Hall term is dominant over the entire temperature range studied. Figure 1(c) displays the resulting estimates for  $\sigma_{xy}^A = \sigma_{xy}$ .

These latter have been used to rewrite the conventional scaling-law equation of state<sup>10</sup> by exploiting the direct proportionality between  $\sigma_{xy}^A$  and M established above. Viz., such proportionality enables the usual Arrott-Noakes/scaling equation of state<sup>21</sup> to be expressed in terms of the anomalous Hall conductivity  $\sigma_{xy}^A$ , i.e.,

$$\left(\frac{\sigma_{xy}^A}{\sigma_1}\right)^{1/\beta} = \frac{T_C - T}{T_C} + \left(\frac{H}{\sigma_{xy}^A}\right)^{1/\gamma} \tag{3}$$

in which  $\sigma_1$  is a material specific constant. The critical exponents appropriate for the corresponding transition are those which then linearize the *anomalous Hall conductivity*—field data plotted in the above form. As Fig. 2(a) shows conclusively, "conventional" Arrott plots of these data, viz.,  $H/\sigma_{xy}^A$  versus  $(\sigma_{xy}^A)^2$ , are indeed linear, a result that demonstrates the applicability of mean-field model exponents [ $\beta$ =0.5,  $\delta$ =3, and  $\gamma$ =1 (Ref. 10)], with the estimate for the ordering temperature being  $T_C$ =61.5±0.5 K [from the critical anomalous Hall conductivity  $\sigma_{xy}^A(T_C)$ ]. This value for  $T_C$  confirms indirectly—through a comparison with the  $T_C/x$  versus *n* phase diagram (*x* being the Mn doping level)<sup>5,8</sup>—that the nominal and actual compositions are very close.

The self-consistency of these exponent estimates was checked as follows. The same replacement of M by  $\sigma_{xy}^A$  in the usual scaling-law equation of state<sup>10,21</sup> leads to

$$\sigma_{xy}^{A}(H,t) = |t|^{\beta} F_{\pm} \left(\frac{H}{|t|^{\gamma+\beta}}\right). \tag{4}$$

 $F_{\pm}$  being an (unspecified) scaling function. Equation (6) then leads to a set of power-law dependences close to  $T_C$ ,<sup>10,18</sup> which here read: for the *spontaneous anomalous Hall conductivity*  $\sigma_{xy-Spon}^A$  (H=0,  $t=|T-T_C|/T_C$ ),

$$\sigma^{A}_{xv-Spon}(0,t) \propto t^{\beta}(T < T_{C}).$$
(5)

Along the critical isotherm  $(T=T_C, t=0)$ , the field dependence of the anomalous Hall conductivity becomes

$$\sigma_{xy}^{A}(H,T=T_{C}) \propto H^{1/\delta}$$
(6)

while for the quantity  $H/\sigma_{xy}^A$ , proportional to the inverse initial susceptibility, one obtains



FIG. 2. (Color online) Unconventional critical analysis based on the anomalous Hall conductivity  $\sigma_{xy}^A$ . (a) The equivalent "Arrott plots"— $H/\sigma_{xy}^A$  vs  $(\sigma_{xy}^A)^2$ ; the straight line passing through the origin yields the critical temperature  $T_C=61.5\pm0.5$  K. (b)  $\sigma_{xy}^A$  vs H along the critical isotherm  $(T_C)$ : inset; these same data replotted on the double-logarithmic scale, the slope of the resulting straight line yielding  $\delta = 2.98 \pm 0.02$ . (c) The spontaneous anomalous Hall conductivity  $\sigma_{xy}^A$ -Spon plotted against temperature: inset;  $\sigma_{xy-Spon}^A$  vs t on a double-logarithmic scale, the slope of the straight line drawn yielding  $\beta = 0.49 \pm 0.02$ . (d)  $H/\sigma_{xy}^A$  plotted against temperature: inset;  $H/\sigma_{xy}^A$  vs t on a double-logarithmic scale, the slope of the straight line drawn yieldis  $\gamma = 1.02 \pm 0.03$ .

$$H/\sigma_{xy}^A \propto t^{\gamma}(T > T_C). \tag{7}$$

The intercepts of the linearized plots of Fig. 2(a) on the abscissa,  $[(\sigma_{xy}^A/\sigma_1)^{1/\beta}]$ , i.e., at H=0, yield the corresponding "spontaneous" anomalous Hall conductivity  $\sigma_{xy-Spon}^A$ , equivalent to the spontaneous magnetization while those on the ordinate axis  $[(H/\sigma_{xy}^A)^{1/\gamma}]$  yield a quantity proportional to the (inverse) initial susceptibility  $[1/\chi_i(T) = (\partial H/\partial M)_{H=0}]$ . The isotherm that passing through the origin [the "critical" anomalous Hall conductivity isotherm,  $\sigma_{xy}^A(T_C)]$  yields the ordering temperature  $T_C$  (the temperature at which  $\sigma_{xy-Spon}^A$  first emerges).

Self-consistency is then achieved by applying Eqs. (5)–(7) to the appropriate intercept values so estimated and substituting the exponents values deduced from them back into Eq. (3) and repeating this process—with *small* adjustments to  $T_c$ —until the Arrott plots and the ensuing power laws yield the same exponent values. The detailed confirmation of these exponent assignments are provided in Figs. 2(b)–2(d), particularly their double logarithmic inserts testing the power-law predictions and yielding  $\beta$ =0.49±0.02,  $\gamma$ =1.02±0.03, and the equation of state critical exponent  $\delta$ =2.98±0.02 (for 2 kOe < H < 70 kOe). These exponent estimates agree,

within experimental uncertainty, with mean-field model values and thus satisfy the Widom relation  $\gamma = \beta(\delta - 1)$ .<sup>10</sup>

A finally comprehensive assessment of the applicability of mean-field exponents to the anomalous Hall conductivity  $\sigma_{xy}^A$  data estimates so obtained for Ga<sub>0.98</sub>Mn<sub>0.02</sub>As is provided by Fig. 3. This figure demonstrates unequivocally that the data in Fig. 2(a) can be scaled onto two branches, one for data below ( $F_-$ ) and the other for data above  $T_C$  ( $F_+$ ), using the choice of variables suggested by Eq. (4) and mean-field exponents; these branches merge as the temperature approaches  $T_C$  (i.e., the reduced temperature *t* approaching zero).

Such a result has interesting consequences. Whereas the critical behavior for localized spins coupled via short-ranged (Heisenberg) interactions in insulators is determined by the space/lattice dimensionality, d, and the order parameter/spin dimensionality, k, the corresponding behavior in metallic systems is markedly different. For the latter, renormalization-group calculations<sup>9</sup> predict that for long-ranged attractive spin-spin interactions [J(r)] decaying with distance r as  $J(r) \approx r^{-(d+k)}$ , mean-field behavior occurs for  $d/2 \le k \le 3/2$  [i.e., if, in three dimensions (d=3), J(r) decreases with r more slowly than  $r^{-4.5}$ , then k=1.5]. The above exponent values clearly satisfy the Widom relationship  $\gamma = \beta(\delta - 1)$ , and when used in conjunction with this latter discussion and



FIG. 3. (Color online) Comprehensive scaling plot for  $\sigma_{xy}^A$  using the critical exponents and  $T_C$  listed above. The  $\log_{10}(\sigma_{xy}^A/t^\beta)$  vs  $\log_{10}(H/t^{\beta\delta})$  plot demonstrates the self-consistent determination of  $T_C$  and critical exponents  $\gamma$ ,  $\beta$ , and  $\delta$ . The upper branch corresponds to data below  $T_C$  and the lower branch to data above  $T_C$ .

 $\gamma = (2 - \eta)/k$  yields a value of 0.5 for the pair-correlation critical exponent  $\eta$ .<sup>9,10</sup> In contrast, for k > 2, short-ranged critical behavior ensues, while in the intermediate regime,  $d/2 \le k \le 2$ , the critical exponents depend on the specific value for *k*. The immediate inference drawn from the present data is that, in Ga<sub>0.98</sub>Mn<sub>0.02</sub>As,  $(d+k) \le 4.5$ .

That this result may not hold at all Mn doping levels is

not surprising; in particular, it is anticipated at higher doping levels where the hole-mediated exchange is weaker and the probability of near-neighbor superexchange antiferromagnetic Mn-Mn interactions is increased.<sup>1</sup> Indeed, mean-field exponents do not linearize the equivalent plots—so-called Arrott plots—at x=0.053 (Ref. 3) and x=0.06 (Ref. 22); the latter exhibit curvature, unlike those displayed above, indicating a crossover to a different universality class. This result is consistent with the predicted suppression of  $T_C$  due to correlated Mn-moment fluctuations within the mean-field model.<sup>1</sup>

While a more extensive series of experiments covering a wider range of Mn doping would enable such crossover effects to be investigated in more detail, the importance of the present study is that it demonstrates: first, that accurate exponent values can be extracted from magnetotransport measurements (specifically the anomalous Hall conductivity), admitting its use in a range of other situations, including lowdimensional spintronic devices where conventional techniques are compromised and second, in the case of low Mn doped metallic Ga<sub>0.98</sub>Mn<sub>0.02</sub>As, these exponents are mean field, thus validating directly the frequently adopted theoretical approach to ferromagnetic (III, Mn)V semiconductors.

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- <sup>1</sup>T. Jungwirth, J. Sinova, J. Masek, J. Kucera, and A. H. Mac-Donald, Rev. Mod. Phys. **78**, 809 (2006).
- <sup>2</sup>H. Ohno, A. Shen, F. Matsukura, A. Oiwa, A. Endo, S. Katsumoto, and Y. Iye, Appl. Phys. Lett. **69**, 363 (1996).
- <sup>3</sup>H. Ohno, Science **281**, 951 (1998).
- <sup>4</sup>F. Matsukura, H. Ohno, A. Shen, and Y. Sugawara, Phys. Rev. B **57**, R2037 (1998).
- <sup>5</sup>J. König, H.-H. Lin, and A. H. MacDonald, Phys. Rev. Lett. **84**, 5628 (2000).
- <sup>6</sup>T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science **287**, 1019 (2000).
- <sup>7</sup>A. H. MacDonald, P. Schiffer, and N. Samarth, Nature Mater. **4**, 195 (2005).
- <sup>8</sup> V. Novák, K. Olejnik, J. Wunderlich, M. Cukr, K. Vyborny, A. W. Rushforth, K. W. Edmonds, R. P. Campion, B. L. Gallagher, J. Sinova, and T. Jungwirth. Phys. Rev. Lett. **101**, 077201 (2008).
- <sup>9</sup>M. E. Fisher, S.-K. Ma, and B. G. Nickel, Phys. Rev. Lett. **29**, 917 (1972).
- <sup>10</sup>H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Clarendon, Oxford, 1971).
- <sup>11</sup>C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972).
- <sup>12</sup>See, for example, Y. Taguchi, Y. Oohara, H. Yoshizawa, N. Nagaosa, and Y. Tokura, Science **291**, 2573 (2001); Z. Fang, N.

Nagaosa, Kei S. Takahashi, A. Asamitsu, R. Mathieu, T. Ogasawara, H. Yamada, M. Kawasaki, Y. Tokura, and K. Terakura, *ibid.* **302**, 92 (2003); W.-L. Lee, S. Watauchi, V. L. Miller, R. J. Cava, and N. P. Ong, *ibid.* **303**, 1647 (2004); S. Paschen, T. Lühmann, S. Wirth, P. Gegenwart, O. Trovarelli, C. Geibel, F. Steglich, P. Coleman, and Q. Si, Nature (London) **432**, 881 (2004); T. Miyasato, N. Abe, T. Fujii, A. Asamitsu, S. Onoda, Y. Onose, N. Nagaosa, and Y. Tokura, Phys. Rev. Lett. **99**, 086602 (2007); S. Onoda, N. Sugimoto, and N. Nagaosa, *ibid.* **97**, 126602 (2006).

- <sup>13</sup>N. Nagaosa, J. Phys. Soc. Jpn. **75**, 042001 (2006); N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong. Rev. Mod. Phys. (to be published).
- <sup>14</sup>M. Lee, Y. Onose, Y. Tokura, and N. P. Ong, Phys. Rev. B 75, 172403 (2007).
- <sup>15</sup>T. Jungwirth, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. 88, 207208 (2002).
- <sup>16</sup>W. Jiang, X. Z. Zhou, and G. Williams (unpublished).
- <sup>17</sup>S. H. Chun, Y. S. Kim, H. K. Choi, I. T. Jeong, W. O. Lee, K. S. Suh, Y. S. Oh, K. H. Kim, Z. G. Khim, J. C. Woo, and Y. D. Park, Phys. Rev. Lett. **98**, 026601 (2007); C. Zeng, Y. Yao, Q. Niu, and H. H. Weitering, *ibid.* **96**, 037204 (2006).
- <sup>18</sup>Y. Pu, D. Chiba, F. Matsukura, H. Ohno, and J. Shi, Phys. Rev. Lett. **101**, 117208 (2008).
- <sup>19</sup>G. Mihály, M. Csontos, S. Bordács, I. Kézsmárki, T. Wojtowicz, X. Liu, B. Jankó, and J. K. Furdyna, Phys. Rev. Lett. 100,

107201 (2008).

- <sup>20</sup>A. Wirthmann, X. Hui, N. Mecking, Y. S. Gui, Tapash Chakraborty, C.-M. Hu, M. Reinwald, C. Schüller, and W. Wegscheider, Appl. Phys. Lett. **92**, 232106 (2008).
- <sup>21</sup>A. Arrott, Phys. Rev. **108**, 1394 (1957); A. Arrott and J. E. Noakes, Phys. Rev. Lett. **19**, 786 (1967); J. H. Zhao, H. P.

Kunkel, X. Z. Zhou, G. Williams, and M. A. Subramanian, *ibid.* **83**, 219 (1999); W. Jiang, X. Z. Zhou, G. Williams, Y. Mukovskii, and K. Glazyrin, *ibid.* **99**, 177203 (2007).

<sup>22</sup>I. Stolichnov, S. W. E. Riester, H. J. Trodahl, N. Setter, A. W. Rushforth, K. W. Edmonds, R. P. Campion, C. T. Foxon, B. L. Gallagher, and T. Jungwirth, Nature Mater. **7**, 464 (2008).