Crossover critical behavior of $Ga_{1-x}Mn_xAs$

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The critical behavior of the ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$ with different concentrations of Mn was experimentally studied by thermal diffusivity measurements in close vicinity of the Curie temperature. Because the inverse of the thermal diffusivity has the same critical behavior as the specific heat, this allowed us to determine the critical exponent α for the samples investigated. As the critical temperature was approached, the value of the critical exponent α, for most of the samples under investigation, showed a clear crossover from the mean-field-like to the Ising-like critical behavior. Investigation of this crossover behavior has enabled us to determine the values of the Ginzburg number and the exchange interaction length in $Ga_{1-x}Mn_xAs$.

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I. INTRODUCTION

The critical behavior in the proximity of the Curie temperature is still one of the central problems in the physics of itinerant ferromagnets. By establishing the universality class for the phase transition, one can obtain information on the range of the exchange interactions determining magnetic order in the system in question. One can then use this information to distinguish between long-range exchange interactions (such as in the mean-field approximation) or short-range interactions (as in the case of the Heisenberg or Ising models).[1]

The ferromagnetic semiconductor $Ga_{1-x}Mn_xAs$ has been studied intensely over the last decade and has become a model system for diluted ferromagnetic semiconductors.[2–4] It is now widely accepted that the ferromagnetism in $Ga_{1-x}Mn_xAs$ arises from the hole-mediated exchange interaction between the local magnetic moments of the Mn, and the mean-field-like Zener model has been widely used to describe this system.[3] However, very recently the mean-field-like behavior of $Ga_{1-x}Mn_xAs$ was questioned,[5] since the value of the critical exponent β of the magnetization for samples with effective Mn concentrations of 8 and 10% has been found to be $\beta = 0.407$, which is more consistent with the short-range Heisenberg model for a disordered magnetic system. From this result, the authors concluded that the assumption of indirect exchange by free carriers as the mechanism of ferromagnetic ordering in $Ga_{1-x}Mn_xAs$ with high concentrations of Mn is questionable. Also, the Curie point singularity in the temperature derivative of resistivity $d\rho/dT$ in $Ga_{1-x}Mn_xAs$ with nominal Mn concentration ranging from 4.5 to 12.5% has been recently investigated.[6] By using the similarity between the critical behaviors of $d\rho/dt$, where $t = |T/T_c - 1|$ is the reduced temperature and the specific heat for metallic ferromagnets,[7] the critical exponent α of the specific heat has been estimated from the $\log(d\rho/dt)$ vs $\log(t)$ plots. All data for various Mn concentrations in these plots collapsed into one common temperature dependence for $T < T_C$ and into another common dependence for $T > T_C$. However, no clear power-law behavior of $d\rho/dt$ was obtained for either side of the transition. At the same time, the temperature dependence of the magnetization for a sample with $T_C = 185$ K revealed a power-law dependence with an approximate exponent $\beta =$ 0.3–0.4, consistent with either the Heisenberg or the Ising behavior. On the other hand, very recently a study of the critical behavior of $Ga_{1-x}Mn_xAs$ was carried out using specific heat measurements.[8] The value of the specific heat critical exponent α for $Ga_{1-x}Mn_xAs$ with Mn concentrations of 1.6% was shown to be close to the critical exponent $\alpha \approx 0.11$ for the three-dimensional (3D) Ising model. In contrast, for $Ga_{1-x}Mn_xAs$ with a Mn concentration of 2.6%, the critical behavior appears to be well described by the mean-field, including the 3D Gaussian fluctuations, approach.

In this work, we present an experimental study of the critical behavior of $Ga_{1-x}Mn_xAs$ with different concentrations of Mn (from 2 to 10%), using thermal diffusivity measurements. One should note that the thermal diffusivity D is related to the specific heat through the equation $C = K/\rho D$ (where ρ is the density and K is the thermal conductivity). Since the thermal conductivity of our GaMnAs samples does not show any critical behavior near the Curie temperature, the inverse of the thermal diffusivity has the same critical behavior as the specific heat.[9]

II. EXPERIMENTAL DETAILS

The $Ga_{1-x}Mn_xAs$ layers with different Mn concentrations were grown on semi-insulating (001) GaAs substrates by using molecular beam epitaxy (MBE). Epilayers with thickness about 60 nm were grown at a low temperature of 270 °C using different temperatures of the Mn source. No postgrowth thermal annealing was performed. The Mn concentration in the layers was estimated from x-ray diffraction measurements and was additionally confirmed by x-ray microanalysis. The temperature dependence of magnetization was measured by a superconducting quantum interference device (SQUID) magnetometer. The thermal diffusivity was measured by using a photothermal method.[10] The back side of the sample was kept in thermal contact with a thin $LiTaO_3$ pyroelectric transducer. The front sample surface was blackened with a thin layer

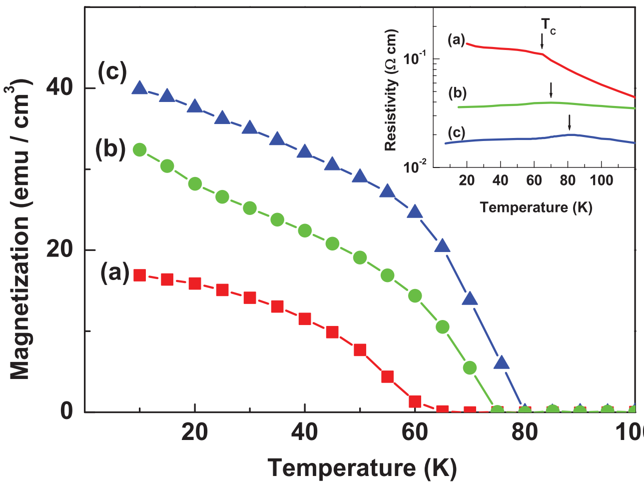

FIG. 1. (Color online) Temperature dependence of the magnetization for the $Ga_{1-x}Mn_xAs$ with (a) 2%, (b) 3%, and (c) 6% of Mn measured in a magnetic field of 10 Oe. Inset: Temperature dependence of the resistivity for the same of $Ga_{1-x}Mn_xAs$ samples measured at a zero magnetic field.

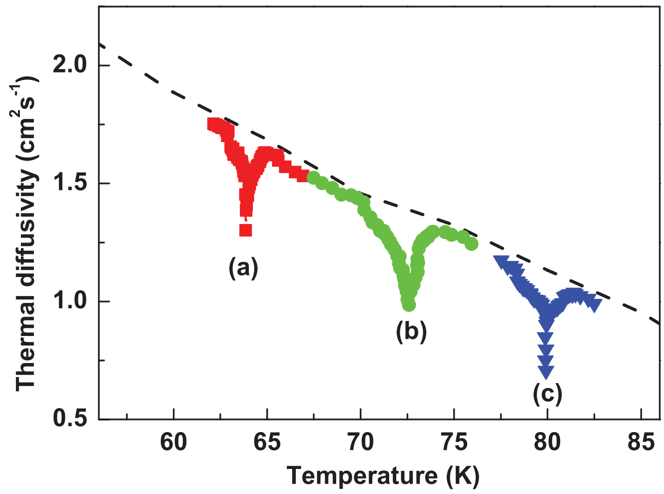

FIG. 2. (Color online) Temperature dependence of the thermal diffusivity for the $Ga_{1-x}Mn_xAs$ with (a) 2%, (b) 3%, and (c) 6% of Mn. The thermal diffusivity of the GaAs substrate is shown by the dashed line.

of carbon black and was heated by an optically modulated He-Ne laser. The modulation frequency was about 3 kHz. At this frequency, the thermal diffusion length is much smaller than the total thickness of the samples (GaMnAs film + GaAs substrate; ~300 μm). This ensures that the investigated samples were thermally thick. The photopyroelectric signal phase was detected by a lock-in amplifier. The sample thermal diffusivity was then calculated by using the total signal phase shift.[10]

III. RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependencies of the magnetization for $Ga_{1-x}Mn_xAs$ samples with three different Mn concentrations. The measurements were conducted at a magnetic field of 10 Oe applied parallel to the sample plane along the easy axis [1$\bar{1}$0] after cooling the samples in a zero magnetic field. From the magnetization curves, the Curie temperatures of the samples have been determined. The Curie temperatures for samples A, B, and C are about 64, 73, and 80 K, respectively. The temperature dependencies of the resistivity for these samples measured in a zero magnetic field are shown in the inset in Fig. 1. Sample A (Mn concentration of 2%) shows an insulating behavior, while samples B and C (Mn concentrations of 3 and 6%, respectively) show metallic behavior. All samples showed a maximum (a rounded cusp) near the Curie temperature T_C, marked by arrows. It should be noted that the resistivity maxima coincide well (within the experimental error) with the corresponding Curie temperatures determined from the magnetization curves.

Figure 2 shows the temperature dependencies of the thermal diffusivity for $Ga_{1-x}Mn_xAs$ samples A, B, and C, and the thermal diffusivity of the GaAs substrate is shown by the dashed line. All samples show a pronounced inverted λ-shaped peak, indicative of a second-order phase transition. The thermal diffusivity peak occurs close to the Curie temperature of the samples, determined from the magnetization measurements, and it is therefore attributed to the ferromagnetic-paramagnetic phase transition. As seen in Fig. 2, the thermal diffusivity peaks are very sharp, which indicates the high crystal quality and homogeneity of the samples being investigated. The thermal diffusivity peak shifts to higher temperatures with increasing Mn concentration. Specifically, these thermal diffusivity peaks for samples A, B, and C occurred at 63.85, 72.59, and 79.91 K, respectively. In order to study the critical behavior of the magnetic phase transition in the proximity of the Curie temperature, we analyzed the inverse of the thermal diffusivity by subtracting the smooth background (dashed line in Fig. 2) as the nonmagnetic contribution of the GaAs substrate and the $Ga_{1-x}Mn_xAs$ layer. The nonmagnetic contribution of the $Ga_{1-x}Mn_xAs$ layers to the thermal diffusivity is expected to be very close to the thermal diffusivity of the GaAs because the Mn concentrations in the investigated samples are relatively low.

Figures 3(a) and 3(b) show plots of the inverse of the thermal diffusivity normalized to the peak maximum as a function of the reduced temperature for the $Ga_{1-x}Mn_xAs$ samples for $T > T_C$ and $T < T_C$, respectively. The expected theoretical value corresponding to the critical exponent of the specific heat $\alpha = 0.11$, predicted by the 3D Ising model, is given by the solid line; and value for $\alpha = 0.5$ corresponding to the mean-field model with 3D Gaussian fluctuations is shown by the dashed line. The contribution of Gaussian fluctuations to the specific heat is given by $\Delta C = C^{\pm}t^{-\alpha}$, where $\alpha = 2 - d/2$, d being the dimensionality.[11,12] In Figs. 3(a) and 3(b) the critical exponent α of the samples investigated clearly shows the crossover from 3D mean-field-like to the 3D Ising-like value as the temperature moves closer to the critical point. Figure 3(c) shows the normalized specific heat data from Figs. 3 and 4 of Ref. 8 for $Ga_{1-x}Mn_xAs$ samples with Mn concentrations of 1.6 and 2.6% vs the reduced temperature, where the data for $T < T_C$ and $T > T_C$ are shown by the filled and empty symbols, respectively. It is seen that the critical exponent α for the sample with 1.6% of Mn shows a clear crossover from 3D Ising-like to 3D mean-field-like

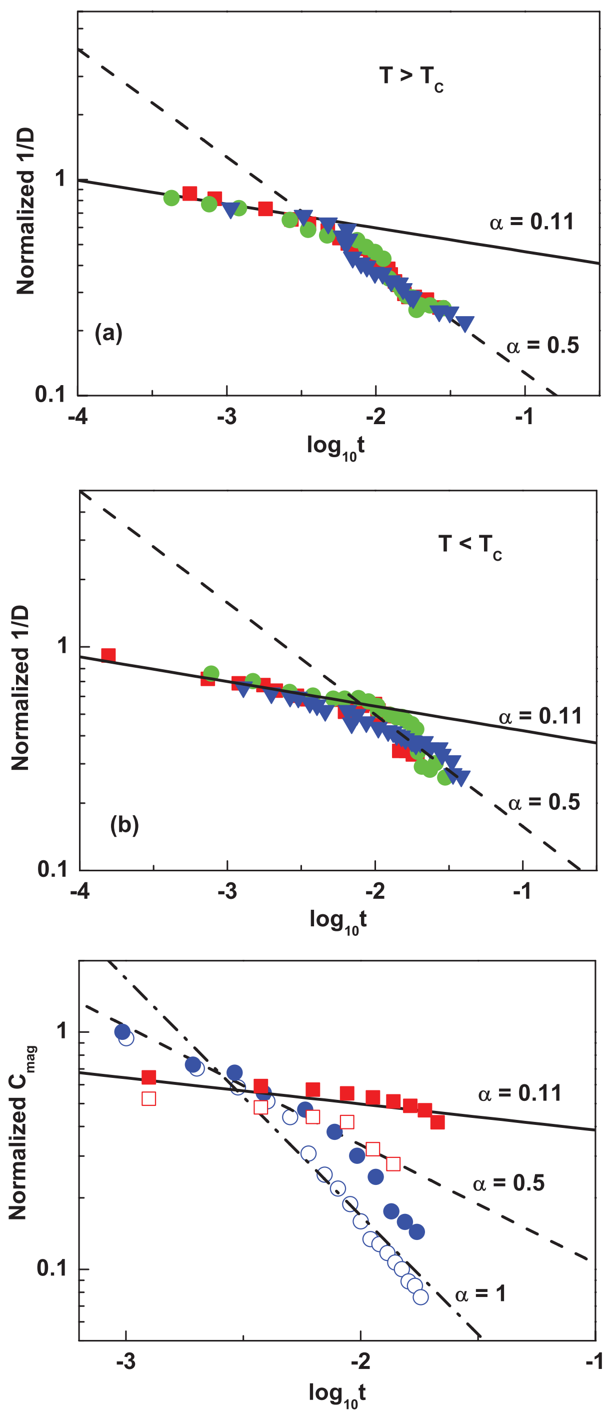

FIG. 3. (Color online) The normalized inverse of the thermal diffusivity vs the reduced temperature for the $Ga_{1-x}Mn_xAs$ samples with 2% (squares), 3% (circles), and 6% (triangles) of Mn for (a) $T > T_C$ and (b) $T < T_C$, respectively. The theoretically expected dependencies for the 3D Ising and the mean-field with the 3D Gaussian fluctuations models are shown by the solid and dashed lines, respectively. (c) The normalized data of the magnetic specific heat for the $Ga_{1-x}Mn_xAs$ samples with 1.6% (squares) and 2.6% (circles) of Mn, derived from experimental data of Ref. 8. The filled and open symbols represent the data for $T < T_C$ and $T > T_C$, respectively. The dashed-dotted line $\alpha = 1$ represents the theoretical dependence of the specific heat for the mean-field model with 2D Gaussian fluctuations.

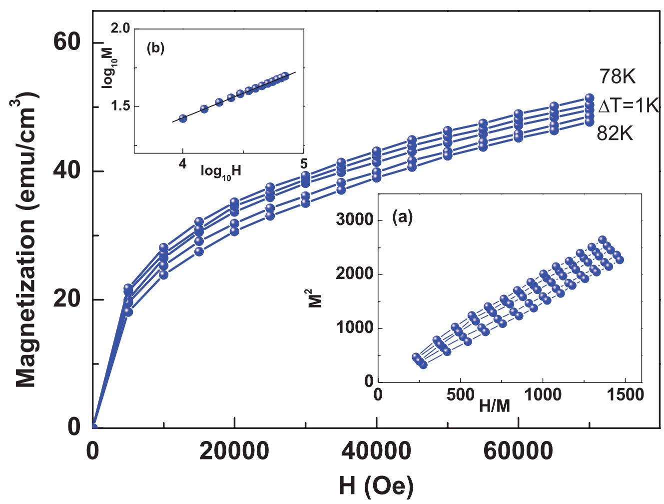

FIG. 4. (Color online) The isothermal magnetization M vs H curves for sample C (6% of Mn) at different temperatures from 78 to 82 K. Only few representative isotherms are shown for clarity. Insets: (a) the M^2 vs H/M (Arrot plot) for different temperatures and (b) the $\log_{10} M$ vs $\log_{10} H$ for $T = 80$ K.

behavior, whereas for the sample with 2.6% of Mn, the critical exponent α does not show any Ising-like behavior for $T < T_C$ and $T > T_C$ until even the smallest value $t = 0.001$ reached in the experiment, but it shows the crossover from 3D to two-dimensional (2D) mean-field-like behavior as the temperature moves away from the critical point. The observation of Ising-like critical behavior in $Ga_{1-x}Mn_xAs$ samples points to the existence of a strong uniaxial magnetic anisotropy in $Ga_{1-x}Mn_xAs$ epitaxial layer, which has already been theoretically and experimentally demonstrated.[13,14]

The critical behavior of a system exhibiting a second-order phase transition is strongly affected by the range of interactions. In the limit of infinite interactions, the system is characterized by the mean-field scaling behavior. However, according to the well-known Ginzburg criterion,[15] the mean-field-like behavior occurs even for finite interaction ranges, sufficiently far away from the critical temperature. The crossover of the critical behavior is governed by the parameter t/G, where G is the so-called Ginzburg number, and the nonclassical critical behavior occurs for $t/G \ll 1$, and that classical critical behavior is expected when $t/G \gg 1$.[16] We determined the value of the Ginzburg number for our samples from Fig. 3, using the midpoint of the crossover from the mean-field to the Ising critical behavior. The value of the Ginzburg number can be also determined from the crossover function obtained numerically by using renormalization group theory.[17] This, however, is beyond the scope of the present paper. As seen from Fig. 3, the Ginzburg number for $T < T_C$ (denoted by G^-) is always larger than for $T > T_C$ (denoted by G^+) for the samples investigated. Although the procedure used for obtaining G cannot provide highly accurate values, and the differences between G^+ and G^- are relatively small, the systematic character of this phenomenon deserves further consideration.

The value of the Ginzburg number in ferromagnets depends on the effective magnetic interaction length R as $G = G_0 R^{-2d/(4-d)}$,[16,18] where d stands for the dimensionality and

R is expressed in units of the lattice constant and hence is dimensionless. As $R \to \infty$, a mean-field-like behavior is observed. However, when R is finite, in the immediate vicinity of the critical temperature $T_{C,}$ one always observes nonclassical critical exponents, which differ from those of the Landau theory. The exchange interaction length can be calculated from the expression given in Ref. 19 which, for the 3D case, reduces to $G = 27/(\pi^4 R^6) \approx 0.277/R^6$.[16] Therefore, from Figs. 3(a) and 3(b) for sample A, we have obtained $R^+ \approx 2$, which represents ~11.3 Å and $R^- \approx 1.75$, which represents 9.9 Å (where the superscripts "+" and "−" correspond to $T > T_C$ and $T < T_C$, respectively. For the $Ga_{1-x}Mn_xAs$ sample with 2.6% of Mn [Fig. 3(c)], both of the Ginzburg numbers below and above T_C are lower than 0.001, and therefore the exchange interaction length for this sample $R^{\pm} > 14.5$ Å.

Near the second-order ferromagnetic phase transition, not only does the specific heat C_p show a power-law dependence on the reduced temperature, but other parameters, such as spontaneous magnetization and magnetic susceptibility, reveal similar behavior with critical exponents β and γ, respectively, and at $T_C M(H) \propto H^{1/\delta}$.[20] The conventional method to determine the critical exponents and critical temperature from the magnetization measurements involves the use of the Arrot plot.[21] Figure 4 shows the isothermal magnetization M vs H curves for sample C measured at different temperatures around $T_C \approx 80$ K. The Arrot plot of the M^2 vs H/M for this sample is shown in the inset of Fig. 4(a). The isothermal curves measured at $\Delta T \geqslant 1$ K from the Curie temperature are linear, which demonstrates a mean-field behavior with $\beta \approx 0.5$ and $\gamma \approx 1$. However, the isothermal curves measured in very close vicinity of the T_C show nonlinear dependence indicating nonmean-field-like behavior. We used a modified Arrot-Noakes plot scheme to determine the critical exponents β and γ from the $M^{1/\beta}$ vs $(H/M^{1/\gamma}$ plot.[22] From this plot the critical exponents $\beta \approx 0.44 \pm 0.02$ and $\gamma \approx 1.06 \pm 0.03$ have been obtained. In order to determine the critical exponent δ, we plot the M vs H using the isothermal curve measured at $T = 80$ K in a double logarithmic scale, shown in the inset of Fig. 4(b). The inverse slope of the $\log_{10} M$ vs $\log_{10} H$ gives $\delta = 3.3 \pm 0.1$. The magnetization measurements of samples A (2% of Mn) and B (3% of Mn) reveal the similar values of the critical exponents within the experimental error. The critical exponents β, γ, and δ obtained from the magnetization experiments are between the 3D Ising model and 3D mean-field values, similar to that observed for $La_{0.75}Sr_{0.25}MnO_3$.[23] From this we infer that the value of $\beta = 0.407$ obtained in Ref. 5 is not due to the Heisenberg short-range exchange interaction in $Ga_{1-x}Mn_xAs$ samples, as was assumed in Ref. 5, but it is rather due to the uncompleted crossover from the mean-field to the Ising-like critical behavior, because the lowest value of the reduced temperature in Ref. 5 was of 0.01. The isotropic Heisenberg-like critical behavior is not consistent with a strong uniaxial magnetic anisotropy observed in $Ga_{1-x}Mn_xAs$ epitaxial layers.

Figure 5 shows the temperature dependence of the thermal diffusivity of the $Ga_{1-x}Mn_xAs$ sample with a Mn concentration of 10%, which was annealed at 160 °C for 24 h in air.[24] The inset in the Figure 5 shows the temperature dependence of the magnetization measured with the magnetic field applied along

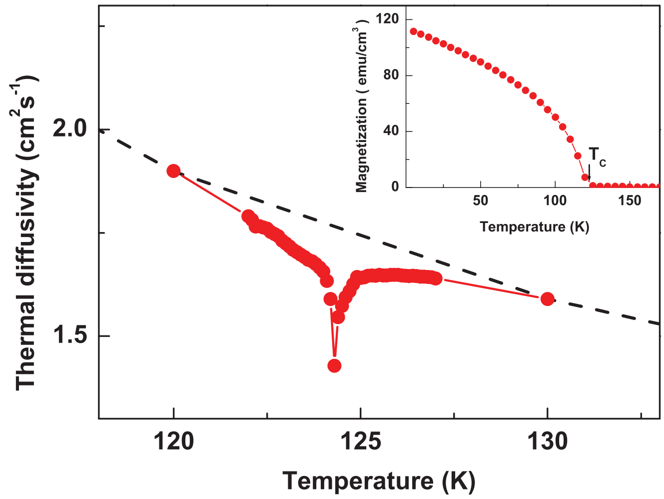

FIG. 5. (Color online) Temperature dependence of the thermal diffusivity for the $Ga_{1-x}Mn_xAs$ with 10% of Mn, annealed at 160 °C for 24 h in air. The thermal diffusivity of the GaAs substrate is shown by dashed line. Inset: Temperature dependence of the magnetization for the annealed sample.

the easy axis [1$\bar{1}$0], indicating a Curie temperature of about 124 K. Figure 6 shows plots of normalized inverse thermal diffusivity vs the reduced temperature above and below the critical temperature. This sample with high Mn concentration demonstrates only the mean-field-like behavior. The critical exponents $\beta \approx 0.5$, $\gamma \approx 1$, and $\delta \approx 3$, determined from the magnetization measurement of this sample (shown in Fig. 7), confirm the mean-field-like behavior. These results indicate that low-temperature annealing of $Ga_{1-x}Mn_xAs$ increases the exchange interaction length.

In Ref. 5 the interaction length was estimated as the mean-free path λ of holes, and for the hole concentration of $p \approx 5 \times 10^{20}$ cm^{-3} the λ is on the order of 5 Å. Such a small value makes the use of the classical approach involving indirect

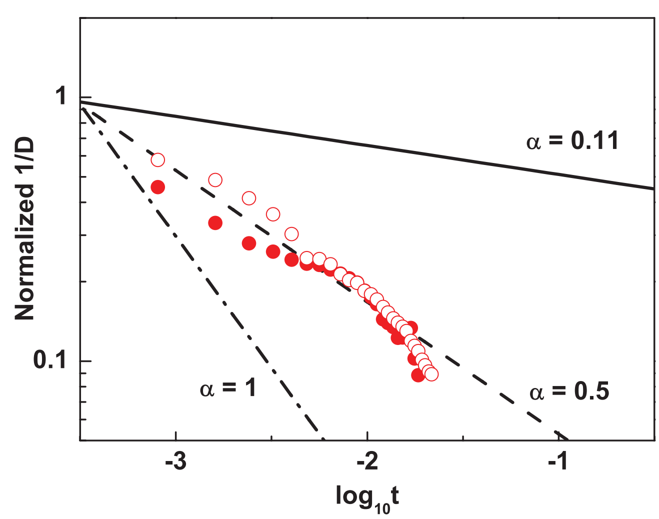

FIG. 6. (Color online) The normalized inverse of the thermal diffusivity vs the reduced temperature for the annealed sample. The filled and open circles represent the data for $T < T_C$ and $T > T_C$, respectively.

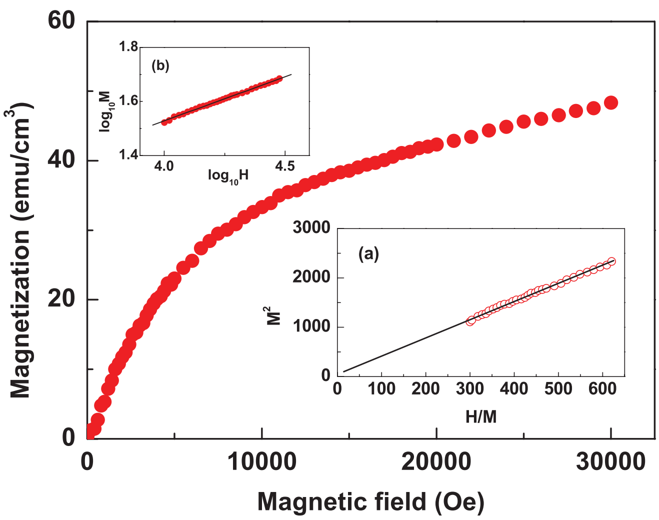

FIG. 7. (Color online) The M vs H dependence for the $Ga_{1-x}Mn_xAs$ with 10% of Mn, annealed at 160 °C for 24 h in air, measured at $T = 124$ K. Insets: (a) the M^2 vs H/M and (b) the $\log_{10}M$ vs $\log_{10}H$ plot.

exchange mechanism by free carriers doubtful. However, the key to the exchange interaction between magnetic ions in $Ga_{1-x}Mn_xAs$ is that it is mediated by spin-polarized holes, and therefore the interaction range should be the spin-diffusion length rather than the mean-free path of the holes. There are several main mechanisms for spin relaxation of carriers in semiconductors: the Elliot-Yafet, the D'yakonov-Perel', and the Bir-Aronov-Pikus.[25] In diluted magnetic semiconductors spin-flip scattering by sp-d exchange interaction should also be considered, particularly because the sp-d exchange is believed to be dominant spin relaxation mechanism in magnetic semiconductors.[25] Hence, although the spin relaxation time of holes in $Ga_{1-x}Mn_xAs$ might be very short, it can still be long compared to the momentum relaxation time.

IV. CONCLUSIONS

In conclusion, we have experimentally studied the critical behavior of $Ga_{1-x}Mn_xAs$ near the Curie temperature by using thermal diffusivity and magnetization measurements. For most of the as-grown samples, we observe a crossover in the behavior of the specific heat critical exponent α from mean-field-like to Ising-like as we approach the Curie temperature from below and above of T_C. The critical exponents β, γ, and δ obtained from the magnetization experiments are between 3D Ising-model and 3D mean-field values. These results show that the external magnetic field used for magnetization measurements suppresses the magnetization fluctuations; therefore, the crossover from the mean-field-like to the Ising-like critical behavior for the critical exponents β, γ, and δ is not completed. The crossover critical behavior demonstrates that the exchange interaction length between magnetic ions in $Ga_{1-x}Mn_xAs$ is finite. However, the as-grown sample with a moderate concentration of Mn (2.6%), as well as the sample with a high concentration of Mn (10%) after the low-temperature annealing, demonstrates the mean-field-like behavior until $t = 10^{-3}$ the smallest value reached in the experiment. These results demonstrate that the exchange interaction length in $Ga_{1-x}Mn_xAs$ might be much larger than of 5Å determined in Ref. 5.

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