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In this paper, we use simultaneous magnetometry and electrical transport measurements to critically examine ways in which the Curie temperature (T_C) values have been determined in studies of dilute magnetic semiconductors. We show that, in sufficiently homogeneous samples, T_C can be accurately determined from remanent magnetization and magnetic susceptibility and from the positions of the peak in the temperature derivative of the resistivity. We also show that the peak of the resistivity does not occur at T_C, as illustrated by a (Ga,Mn)As sample for which the peak of the resistivity is at 213 ± 1 K when T_C is only 178 ± 1 K. © *2014 AIP Publishing LLC*.
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The properties of dilute ferromagnetic semiconductors, such as (Ga,Mn)As, have been extensively investigated in recent years resulting in discoveries of new spin-related physical phenomena and functionalities.[1] Dilute magnetic semiconductors are a very interesting class of materials for which the ratio of the Curie temperature, T_C, to moment density can be very high, but which to date have absolute T_C values below room temperature, precluding their exploitation in spintronics technologies. T_C is a parameter of fundamental importance and internationally many groups are exploring a range of approaches to develop dilute ferromagnetic semiconductors with T_C above room temperature. Several different methods have been used to determine T_C values. Using magnetometry, T_C has been determined from thermo-remanent magnetization measurements,[2,3] or from the temperature-dependence of magnetization under small applied fields. Electrical transport data have also been used to determine T_C. It has been shown that critical fluctuations lead to a peak in the temperature derivative of resistance, $dR_{xx}/dT(T)$, in both dilute ferromagnetic semiconductors[4] and ferromagnetic metals,[5] where in both cases the carrier and moment densities are comparable. For ferromagnetic metals, it has been shown that the peak in $dR_{xx}/dT(T)$ and in the specific heat at T_C have the same functional form, as is predicted.[6] Experimentally it has been shown that the position of the peak in $dR_{xx}/dT(T)$ in ferromagnetic semiconductors can be used to obtain quite accurate values of T_C.[5] In dense moment ferromagnetic semiconductors, the moment density is much larger than the carrier density and it is only long wavelength spin fluctuations which scatter carriers effectively.[7] In this situation, $R_{xx}(T)$ has a peak at T_C.[8] In early studies of dilute ferromagnetic semiconductors, the position of the peak in $R_{xx}(T)$ was mistakenly identified as T_C[9,10] and recently a peak in $R_{xx}(T)$ at 205 K in a (Ga,Mn)As sample was taken to imply an apparent record T_C of 205 K.[11] Finally, T_C values have been obtained from measurements of anomalous Hall resistances, R_{xy}, by assuming a specific relationship between R_{xy} and magnetization.[12]

Close to T_C all the magnetic properties of ferromagnetic materials are determined by critical fluctuations. In the critical region just below a paramagnetic/ferromagnetic phase transition, for zero applied field, the magnetization has power-law behavior given by $M \propto (1 - T/T_C)^{\beta}$.[13] Similarly, just above T_C the low-field susceptibility χ is given by $\chi \propto (T/T_C - 1)^{-\gamma}$. In these expressions, the specific values of the critical exponents, β and γ, depend on the dimensionality and the nature of the order parameter. It follows that

$$\left(\frac{\mathrm{dln}M}{\mathrm{d}T}\right)^{-1} = -\frac{1}{\beta}(T_C - T) \tag{1}$$

and

$$\left(\frac{\mathrm{dln}\chi}{\mathrm{d}T}\right)^{-1} = -\frac{1}{\gamma}(T - T_C). \tag{2}$$

So plots of $(\mathrm{dln}M/\mathrm{d}T)^{-1}$ or $(\mathrm{dln}\chi/\mathrm{d}T)^{-1}$ against T should yield the value of the T_C from the intercept on the T-axis without any assumption about the values of β and γ, and the β and γ values can be obtained from the gradients. These Kouvel-Fisher (KF) plots[14] are established as the best approach for the determination of the critical temperature and exponents from measured data. We will demonstrate elsewhere that the critical exponent values obtained by this method for (Ga,Mn)As are consistent with the three-dimensional (3D)-Heisenberg model,[15] but in this paper our focus is on obtaining T_C values.

The samples studied are 25 nm thick (Ga,Mn)As layers grown on semi-insulating GaAs substrates, with a nominal manganese concentration of 12%. The sample used for the simultaneous magnetometry and transport measurements was annealed for 48 h at 180 °C, a procedure which we have found results in high T_C values.[3]

In previous studies which compared magnetic and transport properties of dilute ferromagnetic semiconductors, separate pieces from the same wafer were used for magnetometry and transport measurements. This is problematic since (i) the processing of samples for transport measurements can change the materials properties and (ii) the two different measuring systems used must have the same accurate temperature calibration. In the present study, transport and magnetometry measurements were performed simultaneously on the same sample within a Quantum Design superconducting quantum interference device (SQUID) magnetometer, using a specially designed ultra-low magnetic moment probe. A simple four-contact design was used to fabricate contact pads onto the sample, which was 4 mm by 5 mm, by standard photolithography and evaporation of a 20 nm layer of titanium and 100 nm layer of gold. The contacts were not annealed and care was taken to avoid heating the sample during evaporation.

The approximate T_C value was established in initial measurements. Remanent magnetization was measured by first field-cooling from room temperature at 300 Oe, and then measuring the sample magnetization while warming in zero applied field. A temperature step size of 0.1 K/min was used in the temperature range around T_C to maintain quasi-static conditions, ensure accurate temperature measurement and maximize the number of data points over the critical region. To obtain low-field susceptibility data, $M(H)$ measurements were taken over a field range of $\pm$200 Oe at intervals of 0.5 K up to 10 K above T_C, and χ was obtained from a linear fit to $M(H)$.

Fig. 1 shows data for the sample before the contacts for the transport measurements were added. Fig. 1(a) shows the remanent magnetization and inverse susceptibility along the $[1\bar{1}0]$ crystalline axis. This (Ga,Mn)As material has strong uniaxial anisotropy and is in a single domain state after field cooling,[16] so the total remanent magnetization is equal to the projection of remanent magnetization along the $[1\bar{1}0]$ easy axis. This can be seen from the comparison in Fig. 1(b) of the measured remanent and field cooled magnetizations which are almost identical apart from close to T_C. It can be seen from Fig. 1(a) that the measured remanent magnetization falls rapidly with increasing temperature and one might estimate a T_C of about 184 K from the data.

The measured inverse susceptibility above T_C shown in Fig. 1(a) clearly does not increase linearly with T-T_C, i.e., it does not show the Curie-Weiss behavior predicted by the mean field model. If one tries to obtain T_C from such a linear fit, the value obtained will be significantly above the true value.

Fig. 1(c) shows the magnetization and susceptibility KF plots. We have applied linear fits to the data close to T_C (within $\sim$2%) but have excluded data points within $\sim$1 K of T_C. The fits give T_C values of 183.7 $\pm$ 0.1 K and 183.9 $\pm$ 0.1 K, respectively. The measured $M(T)$ data clearly deviates from linear behavior close to T_C. The solid line in Fig. 1(a) is the magnetization calculated from the linear fit of the KF plot. This reveals the nature of the deviation of $M(T)$ from power law behavior. The magnetization is suppressed in the region just below T_C and enhanced above it, consistent with a small broadening of the ferromagnetic to paramagnetic transition due to inhomogeneity.[17]

The effect of broadening on the $M(T)$ plot was simulated by numerical evaluation of the following equation, which assumes a Gaussian distribution of T_C:

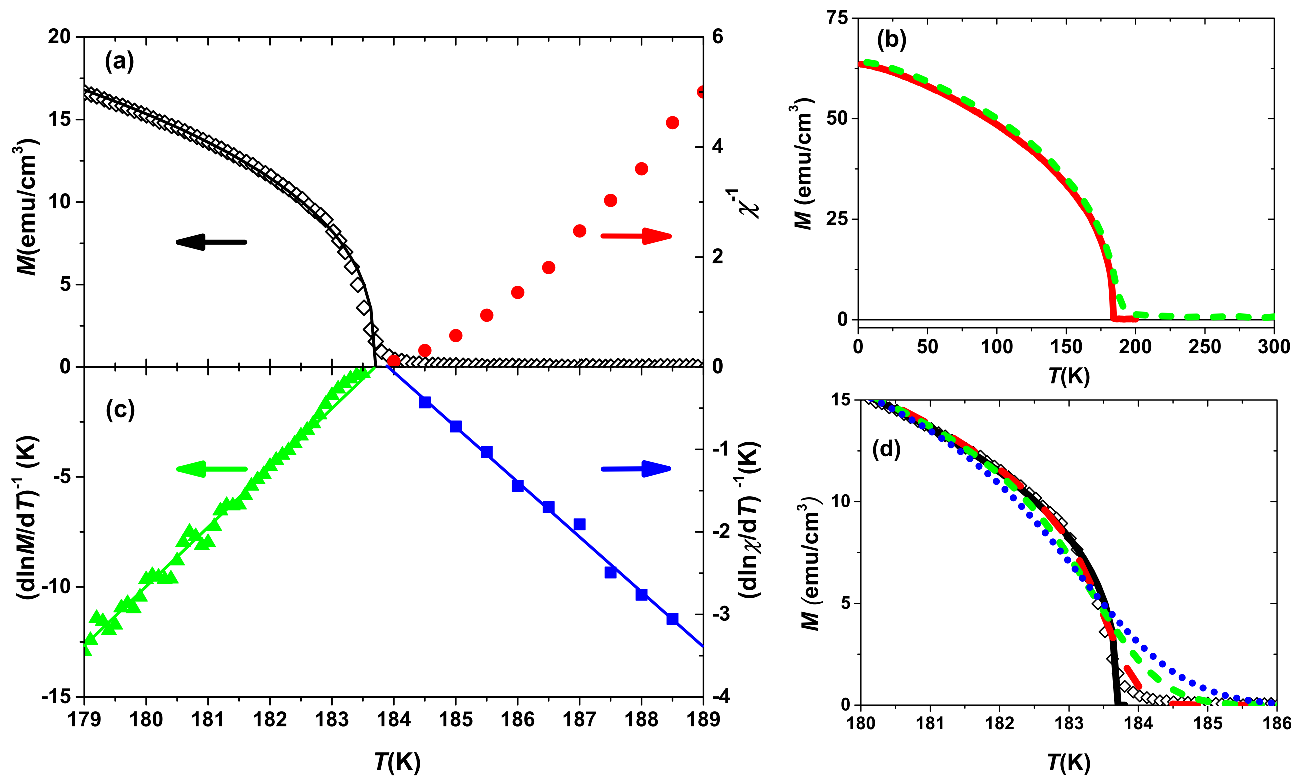

FIG. 1. (a) The projection of the temperature dependent remanent magnetization along the $[1\bar{1}0]$ crystalline axis: measurements (open diamonds) and the behavior from the linear fit to the KF plots for these measurements (solid line) and the measured inverse susceptibility (red dots). (b) The projection of the temperature dependent remanent (red solid line) and field cooled (green short dashes) magnetization along the $[1\bar{1}0]$ crystalline axis. (c) The KF plots of $(\mathrm{dlnM/dT})^{-1}$ and $(\mathrm{dln}\chi/\mathrm{dT})^{-1}$. The lines are the linear fittings within the critical region. (d) The calculated results of M(T) for power law behavior using the critical exponent from the remanent magnetization KF plot with Gaussian broadening ΔT_C of 0 K (black solid line), 0.5 K (red dashes), 1 K (green short dashes), and 1.5 K (blue dots). The diamond points are the experimental data.

$$M(T) = \frac{M_0}{\sqrt{2\pi}\Delta T_C}\int\left(1-\frac{T}{T_C}\right)^{\beta}\exp\left(-\frac{(T_C-T_{C0})^2}{2\Delta T_C^2}\right)dT_C. \tag{3}$$

Fig. 1(d) shows calculated results, using the critical exponent β and the mean critical temperature T_{C0} values obtained from the KF plot, for several values for the Gaussian broadening ΔT_C. This shows that the measured $M(T)$ close to T_C are similar to those expected for a weakly inhomogeneous sample with a broadening of the critical region of order 0.5 K.

Fig. 2 shows the measured $M(T)$, $\rho_{xx}(T)$, and $d\rho_{xx}/dT(T)$ for the sample after the fabrication of contacts for electrical measurements. Despite the precautions taken, the processing has clearly had a small influence on the sample. The T_C has reduced by about 1 K and a shoulder is now present in the magnetization below T_C. This possibly indicates that the material under the contacts has a slightly different T_C from that of the uncovered material. This behavior precludes using KF plots to obtain a high accuracy T_C value. However, it is clear that the position of the peak in $d\rho_{xx}/dT(T)$ is in good agreement with the T_C estimated from the remanence while the peak of $\rho_{xx}(T)$ is ~14 K higher. The peak in $\rho_{xx}(T)$ does not therefore arise from critical fluctuations since it occurs well above T_C.

For an ideal homogeneous ferromagnetic metal or dilute ferromagnetic semiconductor, the temperature dependence of $d\rho_{xx}/dT(T)$ would have a singularity at T_C and would follow that of the specific heat, and can be expressed as[5]

$$\left(\frac{d\rho_{xx}}{dT}\right)_{calc} = \frac{a}{\alpha}\left(|1-T/T_C|^{-\alpha}-1\right)+b, \tag{4}$$

where α is the critical exponent, a and b are constants obtained from the fitting outside of the critical region. However, for inhomogeneous samples, the singularity is replaced with a finite peak, which is shifted to lower temperatures due to the asymmetry in the behavior of $d\rho_{xx}/dT(T)$ above and below T_C.[6] The effect of Gaussian broadening of T_C on the critical behavior of $d\rho_{xx}/dT(T)$ was simulated using

$$\frac{d\rho_{xx}}{dT} = \frac{1}{\sqrt{2\pi}\Delta T_C}\int\left(\frac{d\rho_{xx}}{dT}\right)_{calc}\exp\left(-\frac{(T_C-T_{C0})^2}{2\Delta T_C^2}\right)dT_C. \tag{5}$$

The calculated results with various values of the Gaussian broadening ΔT_C are shown in Fig. 2(c). This again shows that, close to T_C, the measured $d\rho_{xx}/dT(T)$ are similar to those expected for an inhomogeneous sample with a ~0.5 K broadening of the critical region. The calculated results also clearly show the shift of the peak to lower temperatures with increasing Gaussian broadening.

Fig. 3 shows similar results for annealed samples from a different wafer with a nominal Mn concentration of 12%. In this case, the transport and magnetometry measurements were not performed simultaneously. However, the T_C values from magnetometry and from the position of the peak in $d\rho_{xx}/dT(T)$ are in agreement while the peak in $\rho_{xx}(T)$ is 34 K above T_C. For this sample, one would conclude that $T_C = 213 \pm 1$ K (a world record) if the $\rho_{xx}(T)$ peak position is taken as T_C when the actual T_C is only 179 ± 1 K. Moreover, from the peak position of $\rho_{xx}(T)$ one would conclude that the T_C is higher for the sample shown in Fig. 3 than for the sample shown in Fig. 2, when the opposite is true. Therefore, as expected from the theory of scattering from critical fluctuations, the peak in $\rho_{xx}(T)$ is not related to T_C and cannot be used to determine T_C.

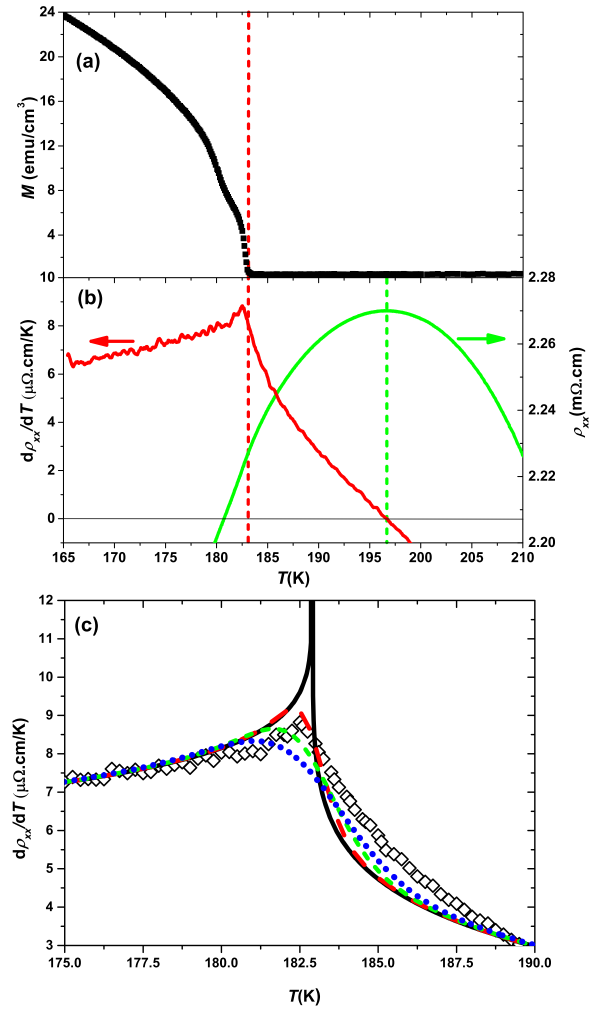

FIG. 2. Simultaneous measurements of (a) the projection of the temperature dependent remanent magnetization along the $[1\bar{1}0]$ easy axis, and (b) the temperature dependence of the resistivity and the temperature derivative of the resistivity ($d\rho_{xx}/dT(T)$), for a 12% Mn 25 nm thick (Ga,Mn)As sample. (c) Calculated results of $d\rho_{xx}/dT(T)$ for power law behavior using the critical exponent from the KF plots with Gaussian broadening ΔT_C of 0 K (black solid line), 0.5 K (red dashes), 1 K (green short dashes), and 1.5 K (blue dots). Diamond points: the experimental data.

The experimental results presented here are for samples which are quite homogeneous with a remanent magnetization which falls very rapidly to zero close to T_C and for which fitting to the data indicates a T_C broadening of order 0.5 K for a T_C of order 180 K. In Fig. 4, we show calculated results showing the effects of larger values of broadening on both $M(T)$ and $d\rho_{xx}/dT(T)$. The values of T_C and ΔT_C used in the calculation are chosen so as to reproduce the behavior seen in Fig. 3 of Ref. 4 in which as-grown samples were subject to a series of annealing steps which successively increase the T_C and the sample homogeneity. With increasing broadening,

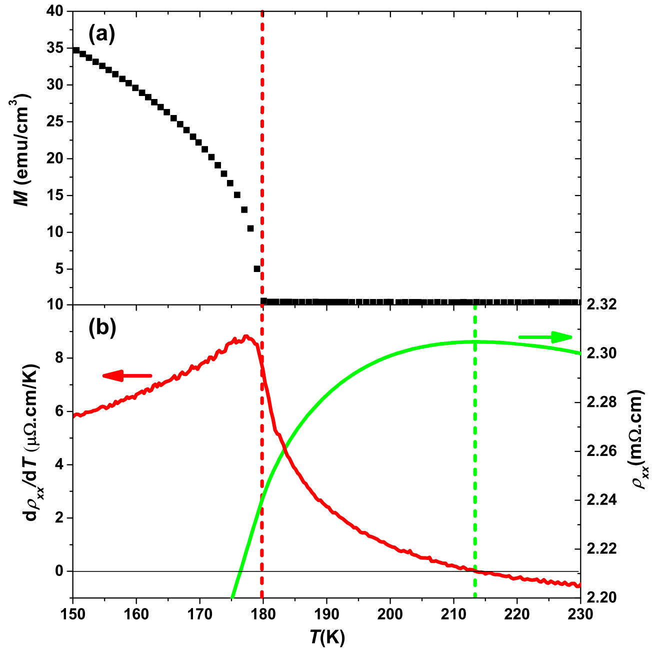

FIG. 3. (a) The projection of the temperature dependent remanent magnetization along the $[1\bar{1}0]$ easy axis for a second 12% Mn 25 nm thick (Ga,Mn)As sample. (b) The temperature dependence of the resistivity and the temperature derivative of the resistivity ($d\rho_{xx}/dT(T)$) from a Hall bar patterned from the same wafer.

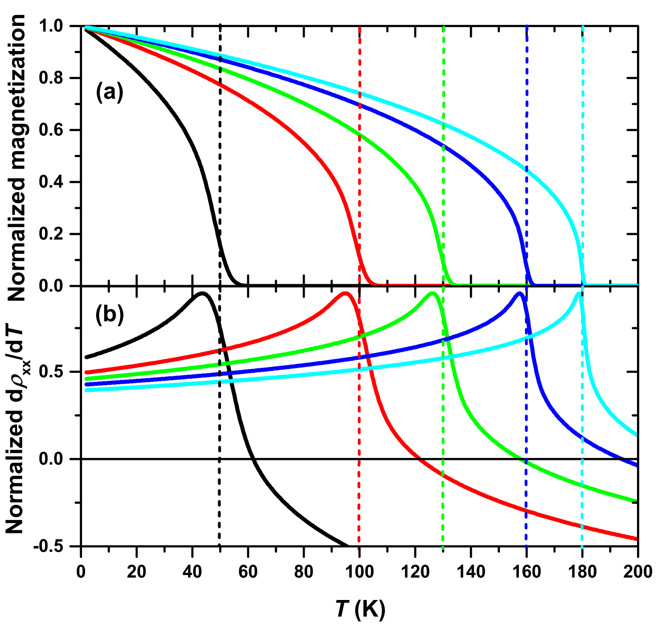

FIG. 4. (a) The calculated results of M(T) for power law behavior with Heisenberg critical exponents with T_C and Gaussian broadening ΔT_C. (b) Calculated results of $d\rho_{xx}/dT(T)$ for the same parameters as (a). The T_C for each of the curves is 50 K, 100 K, 130 K, 160 K, and 180 K, respectively, with ΔT_C equal to 5 K, 4 K, 3 K, 2 K, and 1 K, respectively.

$M(T)$ has an increasing tail extending well above the mean T_C, and it becomes increasingly difficult to estimate T_C from $M(T)$. At the same time, the $d\rho_{xx}/dT(T)$ peak moves below T_C. For the situation of significant inhomogeneity modeled by these calculations, which applies to early studies of (Ga,Mn)As,[9] one can at best make rather imprecise estimates of the mean T_C from the measured $M(T)$ or $d\rho_{xx}/dT(T)$. Note that the behavior of Fig. 4, and Fig. 3 of Ref. 4, is very different from the observed behavior of fully annealed samples over a range of Mn concentrations (Fig. 2 of Ref. 4), for which the peak in $d\rho_{xx}/dT(T)$ remains sharp and close to T_C even when the Mn concentration and T_C are low.

In conclusion, we have shown that KF plots of remanent magnetization or susceptibility can give accurate values of T_C. However, homogeneity limits the temperature range over which such plots show linear behavior and leads to overestimation of T_C if one tries to identify this as "the temperature at which the magnetization goes to zero." We have also shown, by simultaneous magnetometry and transport measurements, that the peak in $d\rho_{xx}/dT(T)$ can be used to obtain an accurate estimate of T_C, though inhomogeneity generally leads to the peak position being slightly below T_C. We have demonstrated that the peak in $\rho_{xx}(T)$ can be far above T_C and cannot be used to estimate T_C values.

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