Specific Heat Study of GaMnAs

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Specific heat was used to study the magnetic phase transition in GaMnAs. Two types of samples were investigated. The sample with a Mn concentration of 1.6% shows an insulating behavior whereas the sample with a Mn concentration of 2.6% is metallic. The temperature dependence of the specific heat for both samples reveals a lambda-shaped peak near the Curie temperature, which indicates a second-order phase transition occurring in these samples. The critical behavior of the specific heat for the GaMnAs samples is consistent with the mean-field behavior with Gaussian fluctuations of the magnetization in the vicinity of $T_C$. © 2010 The Japan Society of Applied Physics

**Ferromagnetic GaMnAs semiconductors have been studied intensely over the last decade and have become a model system for diluted magnetic semiconductors.** ([1](#))[3] It is now widely accepted that the ferromagnetism found in GaMnAs arises from the hole-mediated interaction between the local magnetic moments of the Mn. ([3](#)) However, over the past years, an intense debate has been sparked about the nature of the hole states in this material: are the holes free and reside in a valence band ([6](#)) or they are localized in an impurity band ([2](#))[5]? The study of the critical behavior of the ferromagnets near the Curie temperature is very helpful in understanding their magnetic and electronic properties. By establishing the universality class for the phase transition, information is provided on the range of the exchange interactions. Examples are, a long-range exchange interaction in the case of the mean-field or a short-range interaction in the case of the Heisenberg or Ising models. ([7](#)) Very recently, the Curie point singularity in the temperature derivative of the resistivity in GaMnAs with a Mn concentration ranging from 4.5 to 12.5% has been investigated. ([8](#)) By using the similarity between the critical behaviors of the $d\rho/dT$ and the specific heat for metallic ferromagnets, ([9](#)) the critical exponent $\alpha$ of the specific heat has been estimated from the $\log(d\rho/dt)$ vs $\log(t)$ plots. All of the data sets are distributed between the two common temperature dependencies for $T < T_C$ and $T > T_C$. However, no clear power-law behavior in the $d\rho/dT$ on the either side of the transition has been observed.

In this paper, we present the results of an experimental study of the specific heat near the Curie temperature of GaMnAs having a relatively low concentration of Mn. The temperature dependencies of the resistivity, the magnetization and the specific heat have been observed. To the best of our knowledge no specific heat measurement results for GaMnAs have been previously reported. The GaMnAs layers with low Mn concentrations ($x < 0.03$) were grown on semi-insulating (001) GaAs substrates by using MBE. The epilayers, with a thickness of about 1 μm were grown at the low temperature of 270 °C with different temperatures for the Mn source. The Mn concentration in the layers was estimated using X-ray diffraction measurements and further confirmed by X-ray microanalysis. No post-growth thermal annealing was performed. The hole concentration measured at room temperature for two typical samples with concentrations of Mn at 1.6 and 2.6% were $2.7 \times 10^{19}$ and $4.5 \times 10^{19}$ cm$^{-3}$, respectively. We are aware that the concentration of free holes obtained from Hall measurements for GaMnAs is not rigorously accurate due to the presence of the term arising from the anomalous Hall effect (AHE). However, because of the relatively low Mn concentration in the samples studied, the AHE contribution is expected to be negligible at room temperature. The temperature dependence of the resistivity was conducted by using the LakeShore system equipped with a closed cycle cooling cryostat. The temperature dependence of the magnetization was measured using a superconducting quantum interference device (SQUID) magnetometer. The specific heat was measured by using the $3\omega$ method described elsewhere. ([10](#)) The heating current frequency was 1 MHz and the sweep rate was 100 mK/min.

Figure 1 shows the temperature dependence of the magnetization for the two GaMnAs samples with the Mn concentrations of 1.6% (sample A) and 2.6% (sample B). The measurements were conducted in a magnetic field of 10 Oe applied in parallel to the sample plane after cooling.
In our previous paper the resistivity maximum at the Curie temperature, within experimental errors. The respective $C_T$'s are shown in Fig. 1 by solid arrows. The Curie temperatures for samples A and B are about 40 and 52 K, respectively. The temperature dependence of the resistivity for these samples, measured in a zero magnetic field, is shown in the inset of Fig. 1. It is seen that sample A demonstrates an insulating behavior, while sample B shows a metallic behavior. Both of the samples exhibited a maximum (a rounded cusp) at the Curie temperature $T_C$. It should be noted that the resistivity maximum coincides with the Curie temperature, determined from the magnetization curves, within experimental errors. In our previous paper the resistivity maximum at the GaMnAs Curie temperature was explained by the magneto-impurity model proposed by Nagaev.

Figure 2 shows the temperature dependence of the specific heat $C_p$ for the GaMnAs samples (solid lines) and the GaAs substrate (dashed-dotted line). The specific heat curves of the GaMnAs samples show a pronounced $\lambda$ shaped peak, which indicates the existence of a second-order phase transition within these samples. The specific peak maximum in the GaMnAs samples is located near the Curie temperature, and therefore, is attributed to the ferromagnetic-paramagnetic phase transition. As seen in Fig. 2, with an increase of the manganese concentration the specific heat peak increases in amplitude and shifts to a higher temperature. The specific heat peak maximum for GaMnAs samples A and B was observed to be 39.95 and 51.75 K, respectively.

Figures 3 and 4 show the temperature dependence of the magnetic specific heat $C_{mag}$ for the investigated samples, which was obtained by subtracting the smooth background of the specific heat of the GaAs substrate. The nonmagnetic contribution of the GaMnAs layers to the specific heat is supposed to be very close to the specific heat of the GaAs because the Mn concentration in the samples investigated is relatively low. The critical behavior of the specific heat near the phase transition is described by $C_p = C^\pm t^{-\alpha}$, where $C^\pm$ are the critical amplitudes of the specific heat above (+) and below (−) $T_C$, $t = |T - T_C|/T_C$ is the reduced temperature, and $\alpha$ is the critical exponent of the specific heat. The insets in Figs. 3 and 4 show the plots of the magnetic specific heat vs the reduced temperature using a double logarithmic scale, where $T_C$ is the maximum of the specific heat peak. It is seen, that for the $10^{-3} \leq t \leq 10^{-2}$ regarding the reduced temperature interval close to the $T_C$, the experimental data above and below $T_C$ have a similar slope. For sample A the slope is about 0.09, while for sample B the slope is about 0.5. The value of the critical exponent $\alpha = 0.09$ is close to the critical exponent $\alpha \approx 0.1$ of the three-dimensional (3D) Ising model. This is an unexpected result, because the Ising critical behavior is valid for short-range exchange interactions, while a long-range mean-field-like exchange interaction mediated by free or localized in the impurity band holes is what was expected. However, 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 $\beta$ and $\gamma$, respectively, and at $T_C M(H) \propto H^{1/3}$. Figure 5 shows the $M(H)$ curve for sample A measured at the Curie tempera-
ture of 40 K. The Arrott plot of the $M^2$ vs $H/M$ for this sample is shown in inset (a) of Fig. 5. It is seen that the isothermal curve is linear, which demonstrates the mean-field behavior with $\beta \approx 0.5$ and $\gamma \approx 1$. In order to determine $\delta$, we plot the $M$ vs $H$ using a double logarithmic scale, shown in inset (b) of Fig. 5. The inverse slope of the $\log M$ vs $\log H$ gives $\delta = 3.1$ which is close to the mean-field value of 3. Similar values of the critical exponents $\beta$, $\gamma$, and $\delta$ were also observed for sample B (not shown). The Ising-like critical behavior of the specific heat for sample A is inconsistent with the mean-field values for the magnetization exponents. However, Ising-like critical behavior of the specific heat has been observed for the itinerant ferromagnet of SrRuO$_3$.\cite{14}

explained by the mean-field critical behavior, including the three-dimensional (3D) Gaussian fluctuations. Gaussian fluctuations are associated with the variance in $M((\Delta M^2) = \langle M^2 \rangle - \langle M \rangle^2)$.\cite{13} They occur on a short length scale and do not change the significant mean value of $\langle M \rangle$, therefore, they have no significant effect on the magnetization, but contribute to $C_p$.\cite{14} The contribution of Gaussian fluctuations to the specific heat is given by $\Delta C = C^+ f^{-\omega}$, where $\omega = 2 - d/2$, and $d$ is the dimensionality.\cite{15,16} The amplitude ratio $C^+/C^- = n/z^{1+\omega}$, where $n$ is the number of spin components.\cite{13} With an increasing of the Mn concentration, the contribution of the Gaussian fluctuations to the specific heat of the GaMnAs increases; the critical exponent $\omega \approx 0.5$ is clearly observed for sample B above and below $T_C$ (see the inset of Fig. 4). The value of $C^+/C^- = 0.37$, determined from this experimental plot, is close to the theoretical value of $C^+/C^- = 0.35$ for $n = 1$, which shows the presence of a strong magnetic anisotropy in the GaMnAs samples. The Gaussian fluctuation analysis is valid in the same temperature range as the mean-field theory and hence has the same Ginzburg criterion for validity: $t > (1/32 \pi^2)(k_B/\Delta C\xi^2)^2$, where $\Delta C$ is the specific heat jump at $T_C$ and $\xi$ is the correlation length.\cite{17,18} Taking $\Delta C = 20 J/(\text{mole-K})$ from Fig. 4 and using the smallest $t = 0.001$ in our experiment yield a lower boundary for the correlation length $\xi > 6.9 \text{ Å}$.

Very recently, a multifractal spatial variation of the local density of states (LDOS) near the Fermi energy point has been observed in GaMnAs by using scanning tunneling microscopy.\cite{19} The LDOS distribution shifts away from the Gaussian toward a log-normal distribution with a decreasing Mn concentration. However, log-periodic oscillations in the specific heat of the system with a multifractal distribution is expected,\cite{20} which were not observed in our specific heat measurements. Therefore, our specific heat data support the delocalized carriers model with a Gaussian distribution, especially in the case of metallic GaMnAs.

In conclusion, we performed magnetization, resistivity and specific heat measurements on GaMnAs in order to study the critical behavior of the magnetic phase transition in this material. In a close vicinity to the Curie temperature, the temperature dependence of the resistivity and the specific heat reveal peaks related to the ferromagnetic-paramagnetic phase transition. From a detailed analysis of the specific heat data above and below $T_C$, the value of the critical exponent $\alpha$ has been determined. The critical behavior of the specific heat of the GaMnAs samples is well described by the mean-field, including the Gaussian fluctuations model, which is irrespective of whether the carriers are in the valence band or in the impurity band. That is, even if the carriers are in the impurity band, they are delocalized and the mean field theory still works.

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