

# Magnetic Properties of Layered Itinerant Electron Ferromagnet $\text{Fe}_3\text{GeTe}_2$

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We have successfully synthesized a single-crystal sample of the layered structure ferromagnet  $\text{Fe}_3\text{GeTe}_2$  and present the results of its complete characterization using powder X-ray diffraction (XRD), magnetic susceptibility  $\chi(T)$ , isotherm magnetization  $M(H)$ , specific heat  $C(T)$ , and electrical resistivity  $\rho(T)$  measurements. The rietveld refinement of the powder XRD data confirms that  $\text{Fe}_3\text{GeTe}_2$  crystallizes in the space group  $p6_3/mmc$ . The  $\chi(T)$ ,  $C(T)$ , and  $\rho(T)$  data indicate bulk ferromagnetic ordering at  $T_C = 220$  K with a saturation moment of  $1.625 \mu_B/\text{Fe}$  at zero temperature. The magnetic measurements of the external magnetic field  $H$  applied along the  $c$ -axis and  $ab$ -plane show a large anisotropy in this material. We found that the modified Arrott plot ( $M^4-H/M$ ) rather than the  $M^2-H/M$  plot is obeyed in this material. The generalized Rhodes–Wohlfarth ratio indicates an itinerant magnetism in this material. The spin fluctuation parameters are estimated and the magnetic properties are discussed in the framework of the self-consistent renormalization (SCR) and Takahashi theory of spin fluctuations.

KEYWORDS:  $\text{Fe}_3\text{GeTe}_2$ , itinerant ferromagnet, SCR, Arrott plot

## 1. Introduction

The theoretical and experimental studies of itinerant magnetic materials with layered crystal structure in the frame of the self-consistent renormalization (SCR) theory of spin fluctuations can be traced back to the discovery of cuprate superconductors whose mother compound has two-dimensional characteristics. Compared with Moriya who developed the SCR theory of spin fluctuations for the purely two-dimensional (2D) magnetic system,<sup>1)</sup> Takahashi expanded the SCR theory to the quasi-two-dimensional magnetic system by assuming that the sum of zero-point and thermal spin fluctuations against temperature is nearly conserved.<sup>2)</sup> Ikeda et al. performed a quantitative comparison between the theoretical and experimental results in the  $\text{Sr}_{1-x}\text{Ca}_x\text{Ru}_2\text{O}_7$  system,<sup>3)</sup> and pointed out the possibility of the appearance of quasi-2D weak ferromagnetism in this system on the basis of magnetic measurements. Recently, owing to the discovery of iron-pnictide superconductors, much attention has been paid to the electronic states in 2D magnetic materials, such as the weak itinerant ferromagnets  $\text{LaCoAsO}$ <sup>4–6)</sup> and  $\text{LaCoPO}$ ,<sup>6–8)</sup> which have the same crystal structure as  $\text{LaFeAsO}$ .<sup>9)</sup> Magnetic and nuclear magnetic resonance measurements indicate that 2D spin fluctuations play an important role in these materials, and the magnetic properties were discussed in the framework of the SCR theory. However, the quantitative comparison between the theoretical and experimental results is quite rare, particularly for the magnetic materials with relatively higher Curie temperature.

$\text{Fe}_3\text{GeTe}_2$  is a ternary compound that was first synthesized by Abrikosov et al.<sup>10)</sup> Magnetic measurements show that it is an itinerant ferromagnet with a Curie temperature  $T_C = 230$  K.<sup>11)</sup> The X-ray diffraction (XRD) result indicates that it has a hexagonal crystal structure with the space group  $p6_3/mmc$ , as shown in Fig. 1. Its most pronounced character is the layered  $\text{Fe}_3\text{Ge}$  substructure sandwiched by two layers of Te atoms and a van der Waals gap between adjacent

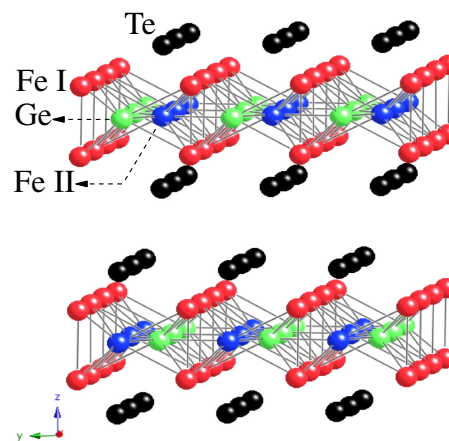


Fig. 1. (Color online) Crystal structure of  $\text{Fe}_3\text{GeTe}_2$ .

Te layers.<sup>11)</sup> Hence, it may be a rare example with a ferromagnetic order in the Fe–Ge–Te system to the best of our knowledge. Anisotropic magnetic properties are strongly expected in this material. In other words,  $\text{Fe}_3\text{GeTe}_2$  provides a good opportunity to perform a quantitative study of the magnetic properties of a quasi-2D itinerant magnetic material.

In this study, we investigate high-quality single-crystal samples of  $\text{Fe}_3\text{GeTe}_2$  by electrical resistivity, specific heat, magnetic susceptibility, and isotherm magnetization measurements in detail. The measurements of the magnetic properties with an external magnetic field applied in the  $ab$ -plane and along the  $c$ -axis indicate that the magnetic properties of this compound show a high anisotropic. Usually, the use of Arrott plots ( $M^2-H/M$ ) is the standard method of determining the Curie temperature  $T_C$ . However, our results suggest that the modified Arrott plot ( $M^4-H/M$ ) rather than the  $M^2-H/M$  plot is obeyed in this compound.  $T_C$  is determined to be 220 K for  $\text{Fe}_3\text{GeTe}_2$  using the modified Arrott plot and electronic resistivity and heat

capacity measurements. Through the quantitative analysis performed on the basis of the Takahashi theory of spin fluctuations, we successfully reproduce the reciprocal magnetic susceptibility above  $T_C$ . Our results indicate that  $\text{Fe}_3\text{GeTe}_2$  is a quasi-2D itinerant ferromagnet.

## 2. Experimental Procedure

We prepared single-crystal samples of  $\text{Fe}_3\text{GeTe}_2$  by the chemical transport method with iodine as the transport agent. High-purity Fe (99.99%), Ge (99.999%), and Te (99.995%) were obtained in a stoichiometric molar ratio of 3 : 1 : 2. All of them were placed in an evacuated quartz tube together with 2 mg/cm<sup>3</sup> iodine. Then, we set up the quartz tube in a temperature gradient of 750/700 °C for one week. Single crystals with a tabular shape in the *ab*-plane were grown on the low-temperature side. The dimensions of a typical sample are  $2 \times 2 \times 0.25$  mm<sup>3</sup>. We determined the molar ratio of the Fe, Ge, and Te concentrations as 3 : 1.02 : 1.98 by energy-dispersive X-ray spectroscopy, which is almost consistent with the nominal molar ratio. The phase purity of the sample was examined by powder XRD analysis using Cu K $\alpha$  radiation.

The magnetization  $M$  and the magnetic susceptibility  $\chi(T) = M/H$ , where  $H$  is the applied magnetic field, were measured in the temperature range of  $2 \leq T \leq 350$  K on a single-crystal sample ( $2.5 \times 2.5 \times 0.2$  mm<sup>3</sup>, 3.0 mg) in a commercial Quantum Design superconducting quantum interference device magnetometer. The dc resistivity  $\rho(T)$  was measured using a standard four-probe technique by applying a current of 5 mA, and the heat capacity  $C_P(T)$  was measured under zero magnetic field. Both  $\rho(T)$  and  $C_P(T)$  measurements were performed using a Quantum Design physical property measurement system. The same crystals were used for these measurements.

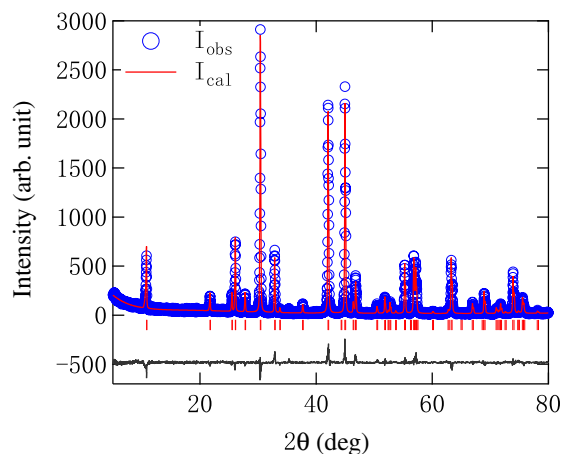
## 3. Results and Discussion

### 3.1 XRD measurements

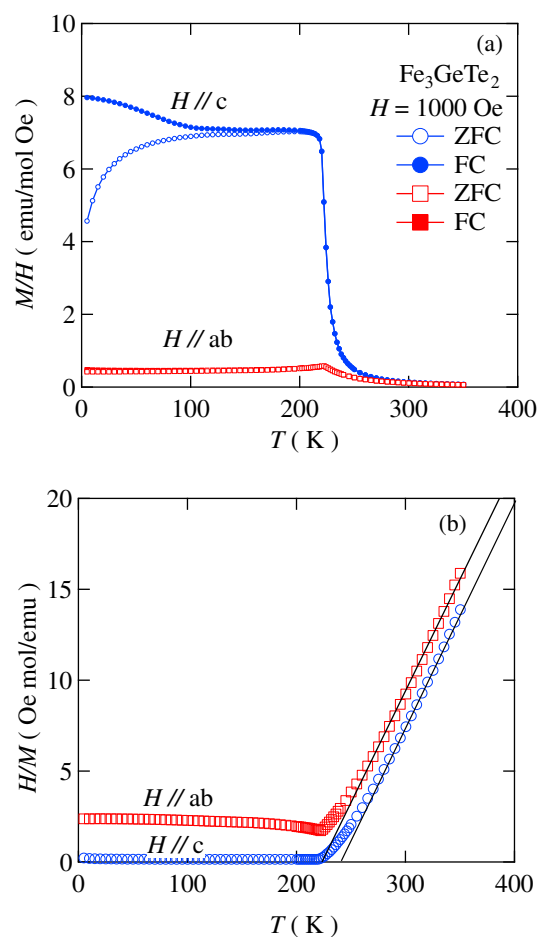
The phase purity of the powdered single-crystal samples was determined using a Rigaku Geigerflex powder X-ray diffractometer with Cu K $\alpha$  radiation ( $\lambda_{\text{av}} = 1.54182$  Å). Rietveld refinements of the data were carried out using the GSAS package.<sup>12,13</sup> Figure 2 shows the Rietveld refinement fit to the X-ray powder diffraction pattern of  $\text{Fe}_3\text{GeTe}_2$ . All the peaks could be indexed and fitted very well on the basis of the hexagonal crystal structure with the  $p6_3/mmc$  space group. The refinement reveals the single-phase nature of the sample without any impurity phase. The obtained lattice parameters are  $a = 4.0299$  Å and  $c = 16.3425$  Å. These values are very close to the reported ones ( $a = 3.991$  Å and  $c = 16.333$  Å).<sup>11</sup>

### 3.2 Magnetic measurements

Figure 3(a) shows the temperature dependence of the magnetic susceptibility  $\chi(T)$  for  $\text{Fe}_3\text{GeTe}_2$  measured with  $H$  ( $= 1000$  Oe) applied parallel to the crystallographic *c*-axis and *ab*-plane, respectively. A rapid increase in  $\chi(T)$  for  $H \parallel ab$  and  $H \parallel c$  at about 220 K suggests the magnetic phase transition at this temperature. The zero-field-cool (ZFC) and field-cool (FC) susceptibilities show significant splitting below 180 K for  $H \parallel c$ , as shown in Fig. 3(a). Such irreversibility is a characteristic behavior of ferromagnetic

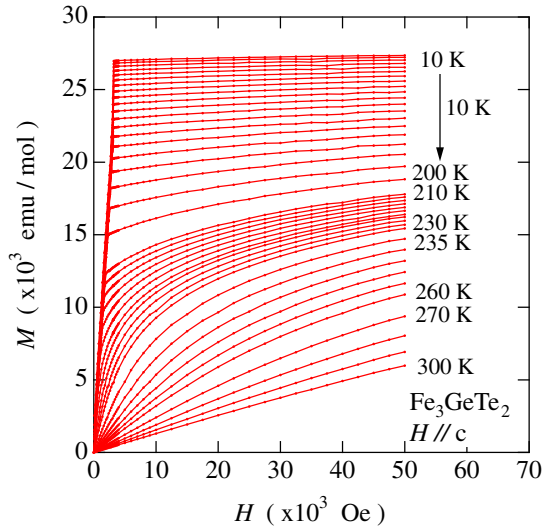


**Fig. 2.** (Color online) X-ray powder diffraction pattern (open circles) measured at room temperature for  $\text{Fe}_3\text{GeTe}_2$ . The solid line represents the Rietveld refinement fit with the  $p6_3/mmc$  space group.



**Fig. 3.** (Color online) (a) Temperature dependence of magnetic susceptibilities of  $\text{Fe}_3\text{GeTe}_2$  measured from 2 to 350 K with the external magnetic field ( $H = 1000$  Oe) applied along the *c*-axis and in the *ab*-plane. (b) Temperature dependence of inverse magnetic susceptibilities for  $\text{Fe}_3\text{GeTe}_2$ . The solid lines indicate the fit with the modified Curie–Weiss law.

compounds. The high-temperature susceptibility shows a weak anisotropy. On the other hand,  $\chi_c$  ( $H \parallel c$ ) is clearly higher than  $\chi_{ab}$  ( $H \parallel ab$ ), e.g.,  $\chi_c/\chi_{ab}$  is about 13.8 at 200 K, indicating the anisotropic behavior in the order state. Furthermore, these results suggest that the easy magnetization direction should be parallel to the *c*-axis.



**Fig. 4.** (Color online) Isothermal magnetization curves for  $\text{Fe}_3\text{GeTe}_2$  measured at various temperatures at magnetic field  $H \parallel c$  of up to 5 T.

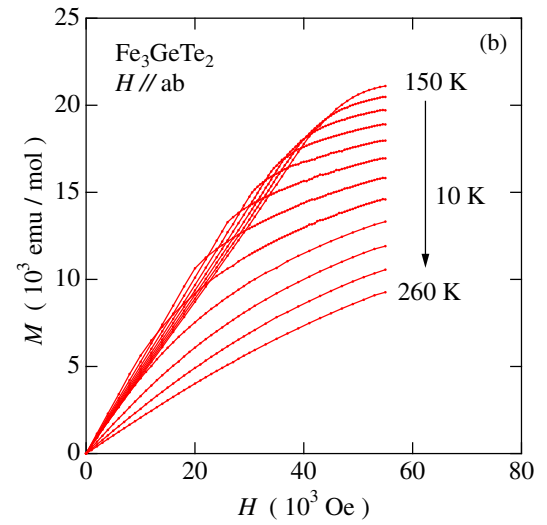
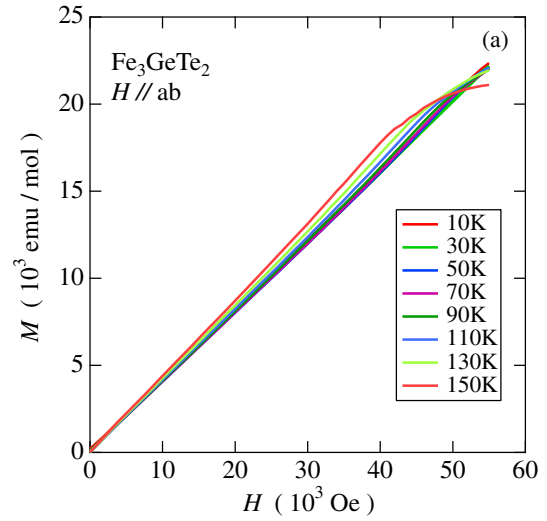
Figure 3(b) shows the corresponding  $T$ -dependence of the inverse susceptibility  $1/\chi$ . Excellent fitting to the data in the high-temperature range was obtained using the modified Curie–Weiss law

$$\chi = \frac{C}{T - \theta} + \chi_0, \quad (1)$$

where  $\chi_0$  is the temperature-independent susceptibility,  $C$  is the Curie–Weiss constant, and  $\theta$  is the Weiss temperature. The values of the parameters obtained from the fitting are  $C = 12.798$  (12.024) emu K/mol,  $\theta = 204.97$  (225.71) K, and  $\chi_0 = -0.026$  (−0.025) emu/mol for  $H \parallel ab$  and  $H \parallel c$ . These results indicate that, at a high  $T$ ,  $\chi(T)$  shows a nearly isotropic paramagnetic behavior. From the fitting parameter, we deduced that the effective moment  $P_{\text{eff}} = 4.7 \mu_B/\text{Fe}$ . A positive  $\theta$  suggests that the ferromagnetic interaction among Fe atoms is dominant between the layers.

Figures 4 and 5 show the isothermal magnetization curves measured for  $\text{Fe}_3\text{GeTe}_2$  in the temperature range from 10 to 300 K with  $H \parallel c$  and  $H \parallel ab$ , respectively. The large anisotropic magnetic properties can be found in the ordered state. For the  $H \parallel c$  shown in Fig. 4,  $M$  shows a saturation behavior at a very low  $H$  of about 3300 Oe. The same behavior is observed in the typical itinerant ferromagnet MnSi that shows a saturation behavior at about 6.2 kOe with  $T = 4.2$  K.<sup>14</sup> With increasing temperature, the saturating magnetization decreases continuously, and  $M$  becomes proportional to  $H$  above 280 K. On the other hand, no saturation behavior but only a concave is observed for  $H \parallel ab$ . For clarity, the  $M$ – $H$  curves for  $H \parallel ab$  with  $T < 150$  K and  $T > 150$  K are plotted in Figs. 5(a) and 5(b), respectively. Note that a saturation tendency can be found at  $H \sim 5$  T for the  $M$ – $H$  curve measured with  $H \parallel ab$  at 10 K as shown in Fig. 5(a). Higher  $H$  measurements are desired to confirm this saturation behavior.

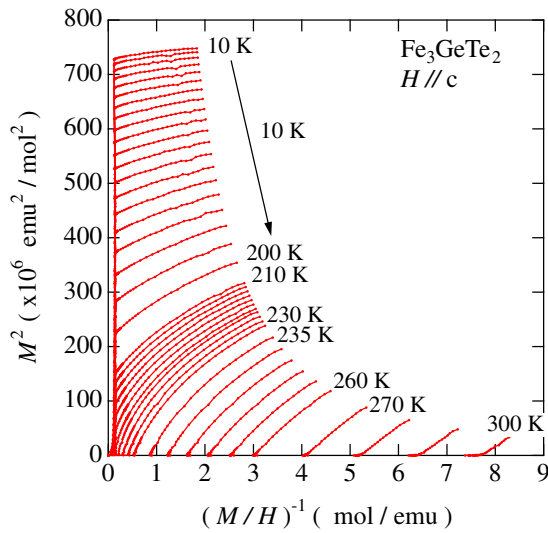
On the basis of the mean-field theory, the Arrott plot ( $M^2$ – $H/M$ ) should be a straight line with an intercept that tends to zero as  $T$  approaches  $T_C$ . Therefore, one can derive the ferromagnetic transition temperature  $T_C$  accurately from



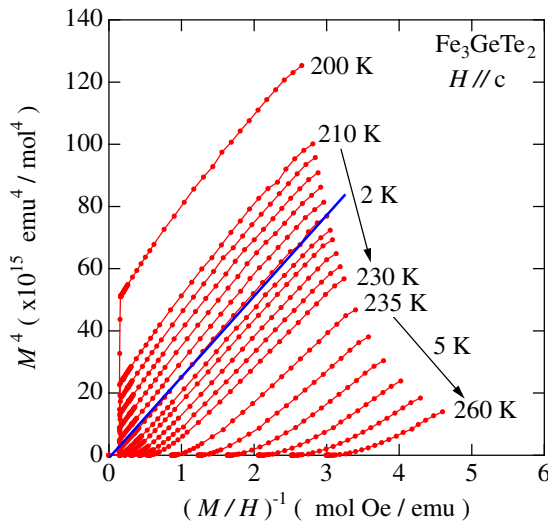
**Fig. 5.** (Color online) Isothermal magnetization curves for  $\text{Fe}_3\text{GeTe}_2$  measured at various temperatures in magnetic field  $H \parallel ab$ -plane of up to 5.5 T. For clarity,  $M$ – $H$  curves for  $H \parallel ab$  with  $T < 150$  K and  $T > 150$  K are plotted in (a) and (b), respectively.

the Arrott plot. The feasibility of this method has been confirmed in some materials, such as  $\text{Y}(\text{Co}_{1-x}\text{Al}_x)_2$ ,<sup>15–17</sup>  $\text{ZrZn}_2$ ,<sup>18</sup>  $\text{Sc}_3\text{In}$ ,<sup>19</sup> and  $\text{Ni}_3\text{Al}$ .<sup>20–22</sup> Figure 6 shows the  $M^2$  vs  $H/M$  plots at various temperatures for  $H \parallel c$ .  $M^2$  does not show a linear relation but a curvature against  $H/M$  at all temperatures, particularly at approximately 220 K. This type of behavior has been observed in some materials, such as MnSi,<sup>23</sup>  $\text{Fe}_x\text{Co}_{1-x}\text{Si}$ ,<sup>24</sup> and  $\text{LaCoAsO}$ .<sup>4</sup> In such cases, the  $M^4$  vs  $H/M$  plot is very useful in determining  $T_C$ , as has been confirmed in these materials. Thus, we replotted the isothermal magnetization curves in the form of  $M^4$  vs  $H/M$ , as shown in Fig. 7. Clearly,  $M^4$  obeys the linear relation against  $H/M$  at approximately 220 K very well.

The linear relationship between  $M^4$ – $H/M$  can be explained by the Takahashi theory, where the field effect of the critical thermal amplitude of spin fluctuations is taken into account. From the  $M^4$ – $H/M$  plot, we determined  $T_C$  as the temperature at which the intercept along the  $M^4$  axis crosses the origin of the coordinate axis.  $T_C$  is determined to be 220 K for  $\text{Fe}_3\text{GeTe}_2$  which is slightly smaller than that reported in Ref. 11 (230 K).

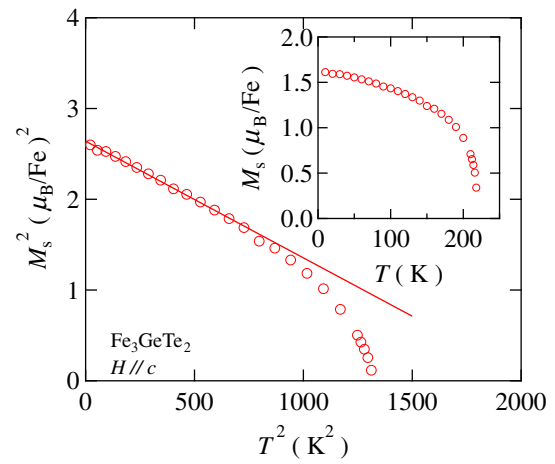


**Fig. 6.** (Color online)  $M^2$  vs  $H/M$  plots (Arrott plot) for  $\text{Fe}_3\text{GeTe}_2$  at various temperatures with  $H \parallel c$ .

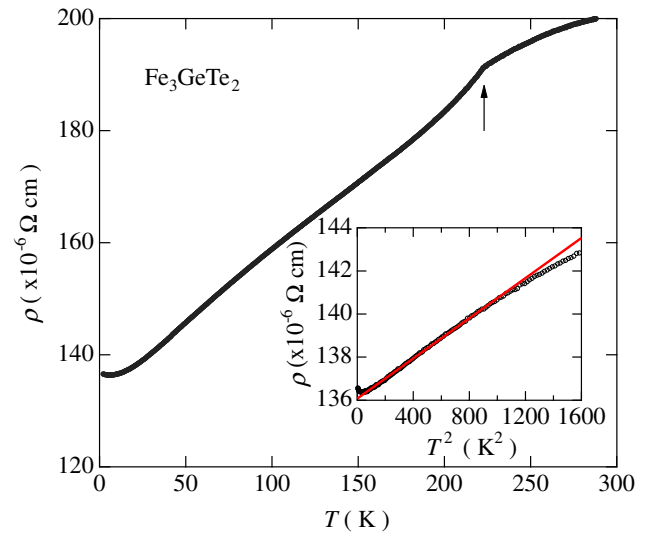


**Fig. 7.** (Color online)  $M^4$  vs  $H/M$  plots for  $\text{Fe}_3\text{GeTe}_2$  at various temperatures with  $H \parallel c$ . At approximately  $T_C$ ,  $M^4$  almost obeys the linear relation against  $H/M$ .

Furthermore, we estimate the spontaneous magnetization  $M_s(T)$  below  $T_C$  from Figs. 6 and 7.  $M_s$  is estimated as the value of the intersection of the extrapolation of the  $M^2$  ( $M^4$ ) curves with the vertical axis. The  $M_s$  value estimated from the  $M^2$  and  $M^4$  curves are almost the same below 150 K, so we only show those estimated from the  $M^2$  curves in this temperature range. The  $M_s$  values between 160 and 220 K are estimated from the  $M^4$ - $H/M$  plot. The obtained temperature dependence of  $M_s$  is shown in the inset of Fig. 8. From the value of the intersection of the natural extrapolation of the  $M_s$  curve with the vertical axis, we obtain the spontaneous magnetization  $P_s = 1.625 \mu_B/\text{Fe}$ . This value is slightly larger than that previously reported ( $1.20 \mu_B/\text{Fe}$ ).<sup>11)</sup> On the other hand,  $T_C = 220$  K, can be considered as the temperature at which  $M_s$  decreases to zero with an increase in temperature. This value is consistent with the  $T_C$  determined from the  $M^4$ - $H/M$  plot.



**Fig. 8.** (Color online)  $M_s^2$ - $T^2$  plot for  $\text{Fe}_3\text{GeTe}_2$ . The inset gives the corresponding  $M_s$ - $T$  plot.



**Fig. 9.** (Color online) Temperature dependence of electrical resistivity of the single crystal of  $\text{Fe}_3\text{GeTe}_2$  with current parallel to the  $ab$ -plane in the temperature range from 2 to 300 K. The arrow indicates the magnetic transition temperature. The inset shows the fit of the low-temperature resistivity data of  $\text{Fe}_3\text{GeTe}_2$  with  $\rho = \rho_0 + AT^2$ .

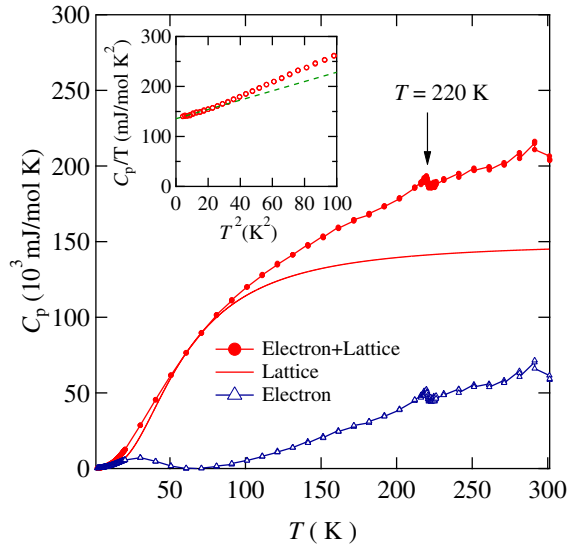
### 3.3 Electrical resistivity

Figure 9 shows the temperature dependence of the electrical resistivity  $\rho(T)$  of the single crystal of  $\text{Fe}_3\text{GeTe}_2$  with current parallel to the  $ab$ -plane in the temperature range from 2 to 300 K. The continuous decrease in  $\rho(T)$  with decreasing temperature suggests the metallic behavior of the compound. The clear anomaly found at  $T = 220$  K indicates the ferromagnetic phase transition, which is consistent with the magnetic measurements. The temperature dependence of resistivity at a low temperature can be well fitted with the power law,  $\rho = \rho_0 + AT^2$  shown in the inset of Fig. 9. We obtained  $\rho_0 = 136.1 \mu\Omega \text{ cm}$  and  $A = 4.66 \times 10^{-9} \mu\Omega/\text{K}^2$ .

### 3.4 Heat capacity

The temperature dependence of the heat capacity  $C_p$  for  $\text{Fe}_3\text{GeTe}_2$  is shown Fig. 10. A clear jump corresponding to the ferromagnetic transition was observed at 220 K, which is





**Fig. 10.** (Color online) Heat capacity  $C_p$  vs temperature  $T$  for single crystal of  $\text{Fe}_3\text{GeTe}_2$  between 2 and 300 K. The arrow shows the magnetic phase transition temperature  $T_C$ . The solid line and triangle indicate the lattice and electron contributions to  $C_p$ . The inset shows the  $C_p(T)/T$  vs  $T^2$  data below  $T = 10$  K. The solid line through the data in the inset is a fit by the expression  $C_p/T = \gamma + \beta T^2$ .

consistent with the magnetic susceptibility and resistivity measurements. The  $C_p$  at room temperature is about 200 J/(mol K), which is larger than the Dulong Petit lattice heat capacity  $C_p = 18R \cong 150$  J/(mol K) as expected for  $\text{Fe}_3\text{GeTe}_2$ ; here,  $R$  is the molar gas constant. This may be due to the magnetic contribution in addition to the lattice contribution.

The inset of Fig. 10 gives the  $C_p(T)/T$  vs  $T^2$  plot for  $\text{Fe}_3\text{GeTe}_2$  at low temperatures. For  $T < 6$  K, the data can be well fitted by the expression

$$C_p(T)/T = \gamma + \beta T^2. \quad (2)$$

Here, the first term is the Sommerfeld electronic specific heat coefficient due to conduction electrons and the second term is the low-temperature limit of the lattice heat capacity. The resultant  $\gamma$  and  $\beta$  are 45.17 mJ/(Fe mol K<sup>2</sup>) and 0.9291 mJ/(mol K<sup>4</sup>), respectively. It is clearly that  $\gamma$  is strongly enhanced by spin fluctuations.

From  $\beta$ , one can estimate the Debye temperature using the expression

$$\theta_D = \left( \frac{12\pi^4 R n}{5\beta} \right)^{1/3}, \quad (3)$$

where  $n$  is the number of atoms per formula unit ( $n = 6$  for our compound).  $\beta$  yields  $\theta_D = 232.36$  K. The calculated lattice contribution to  $C_p$  is shown in Fig. 10 by the solid line. Then, we can deduce the electron contribution to  $C_p$  as shown in Fig. 10 with triangles.

#### 4. Discussion

Usually, the magnetic properties of itinerant materials can be understood in the framework of the Stoner model. However, this model excludes the spin fluctuation effect, leading to some problems remaining unsolved, such as the Curie temperature  $T_C$  and the Curie–Weiss behavior in the high temperature range. Moriya gave the SCR theory of spin

**Table I.** Spin fluctuation parameters of  $\text{Fe}_3\text{GeTe}_2$ .  $T_0$  was deduced from the SCR theory.  $T_A$  was obtained from the slope of the  $M^4$ – $H/M$  plot at 220 K.  $\bar{F}_1$  was estimated from the slope of the Arrott plot at 220 K.  $T_0^*$  and  $\bar{F}_1^*$  were the best fitting parameters. Details can be found in the text.

$T_C$ (K)	$T_0$ (K)	$T_A$ (K)	$\bar{F}_1$ (K)	$T_0^*$ (K)	$\bar{F}_1^*$ (K)
220	2298.77	795.8	96	1455	116

fluctuations in order to take the spin fluctuation effect into account.<sup>1)</sup> Furthermore, Takahashi developed the theory by assuming that the sum of zero-point and thermal spin fluctuations is conserved against temperature; this gives a satisfactory explanation of the  $M^4$  vs  $H/M$  plot, which has been observed in some magnetic materials.<sup>2,23)</sup>

In the Takahashi theory of spin fluctuations, the relationship between  $M^4$  and  $H/M$  is given by

$$M^4 = 1.17 \times 10^{18} \left( \frac{T_C^2}{T_A^3} \right) \left( \frac{H}{M} \right), \quad (4)$$

where  $M$  and  $H$  are in emu/mol and Oe units, respectively.<sup>4)</sup> The parameter  $T_A$  characterizes the dispersion of the spin fluctuation spectrum in wave vector space. Using the linear slope estimated from the  $M^4$ – $H/M$  plot shown in Fig. 7 and  $T_C = 220$  K for  $\text{Fe}_3\text{GeTe}_2$ , we obtained  $T_A = 795.8$  K. On the other hand,  $T_A$  can also be deduced from the  $T^2$ -linear coefficient from the  $M_s^2$ – $T^2$  plot depending on this theory.<sup>20,25,26)</sup> From the linear fitting of the  $M_s^2$ – $T^2$  plot as shown in Fig. 8, we deduce  $T_A = 111.8$  K, which is smaller than the value discussed above.

According to the SCR theory,  $T_C$  can be described as

$$T_C = (60c)^{-3/4} P_s^{3/2} T_A^{3/4} T_0^{1/4}, \quad (5)$$

where  $c = 0.3353 \dots$ ,  $P_s$  is the spontaneous magnetization in  $\mu_B$  unit, and  $T_0$  characterizes the dispersion of the spin fluctuation spectrum in energy space. Using the values of  $T_C$ ,  $P_s$ , and  $T_A$  estimated above, we obtained the characteristic temperature  $T_0 = 2298.7$  K for  $\text{Fe}_3\text{GeTe}_2$ . All the parameters are summarized in Table I.

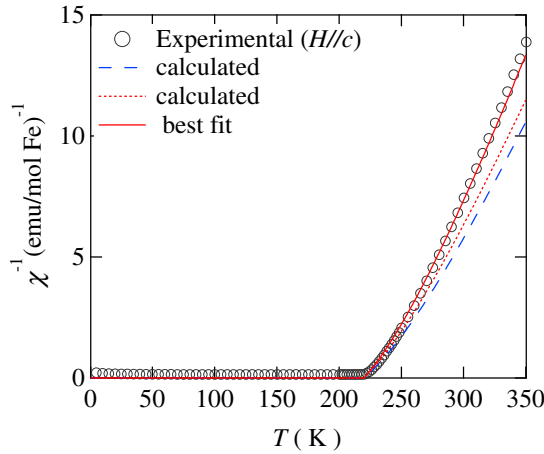
Furthermore, using the above parameters and the mode-mode coupling constant parameter  $\bar{F}_1$ , which is the coefficient of the  $M^4$  term of the Landau expansion of free energy and can be determined from the slope of the Arrott plot ( $\bar{F}_1$  for  $\text{Fe}_3\text{GeTe}_2$  deduced from the Arrott plot is 96 K.), we can calculate the inverse magnetic susceptibility in the frame of the SCR theory for nearly ferromagnetic metals using<sup>23,36)</sup>

$$y = 1 + \eta K_0^2 \int_0^{1/K_0} dz z^3 \left[ \ln u - \frac{1}{2u} - \Psi(u) \right], \quad (6)$$

where  $y = \chi(0)/\chi$ ,  $u = z(z^2 + y)/t$ ,  $K_0^2 = 1/2\alpha T_A \chi(0)$ ,  $\eta = 15\bar{F}_1 T_0 / 2(\alpha T_A)^2$ ,  $t = T/T^*$ ,  $T^* = T_0 K_0^3$ , and  $\Psi(u)$  is the digamma function.<sup>36)</sup> The calculated temperature dependence of the inverse magnetic susceptibility for  $\text{Fe}_3\text{GeTe}_2$  is shown in Fig. 11 with a dashed line. The agreement between theoretical and experimental results is fairly good near  $T_C$ .

On the other hand, in order to modify the discrepancy in the high temperature range, we can also deduce  $T_0$  with the experimental results  $T_A$  and  $\bar{F}_1$  using

$$\bar{F}_1 = \frac{4k_B T_A^2}{15T_0}, \quad (7)$$

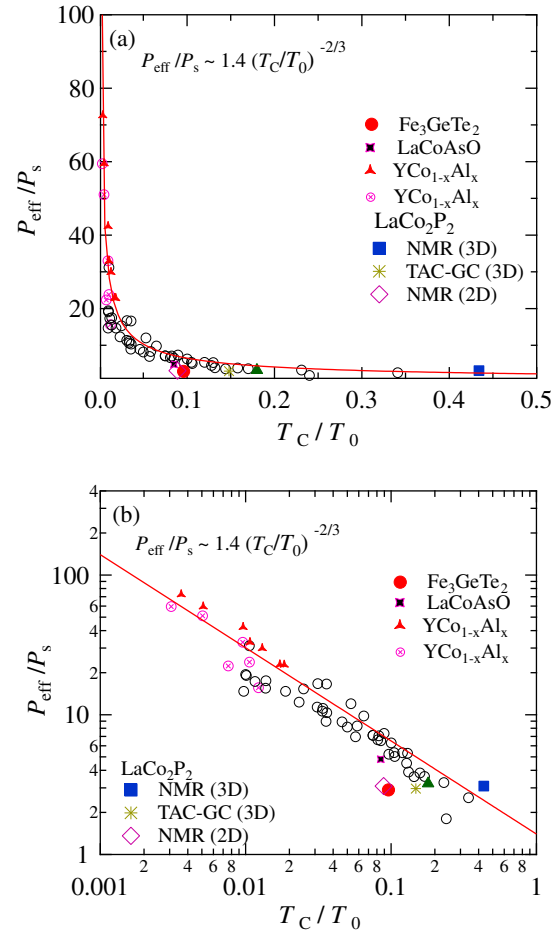


**Fig. 11.** (Color online) Temperature dependence of inverse susceptibility  $\chi^{-1}$ . The dashed line represents the  $\chi^{-1}$  calculated with  $T_0$  determined on the basis of SCR theory. The dotted line indicates the  $\chi^{-1}$  calculated with  $T_0$  deduced using Eq. (7) on the basis of Takahashi theory of spin fluctuations. The solid line shows the best-fitted line for  $\text{Fe}_3\text{GeTe}_2$ .

which can be deduced from the Takahashi theory of spin fluctuations.<sup>2,23,25</sup> The effectiveness of this formula has been confirmed by experiments.<sup>3,20,25,37–39</sup> The calculated value of  $T_0$  is 1760 K. The calculated inverse susceptibility at  $T_0 = 1760$  K,  $T_A$ , and  $\bar{F}_1$  is presented in Fig. 11 using a dotted line. Although the discrepancy is improved, unconformity still exists. Taking into account that  $\bar{F}_1$  has a weak fitting temperature range dependence, we modify  $\bar{F}_1$  and  $T_0$  using Eq. (7), to obtain the best fitting of the inverse susceptibility. The best fitting parameters  $T_0^*$  and  $\bar{F}_1^*$  are indicated in Table I, and the corresponding fitting curve is shown in Fig. 11 by a solid line.

In order to clarify whether a magnetic material belongs to the itinerant or a localized spin system, the Rhodes–Wohlfarth ratio is a very good criterion in the Stoner model, which is defined as  $P_c/P_s$ , where  $P_c(P_c + 2) \equiv P_{\text{eff}}^2$  and  $P_s$  is the spontaneous magnetization in the ground state. In the case of a weak itinerant ferromagnet, the Rhodes–Wohlfarth ratio is much larger than that in the case of the localized spin system  $P_c/P_s \equiv 1$ . The rather large value of  $P_c/P_s \equiv 3.8$  for  $\text{Fe}_3\text{GeTe}_2$  indicates that the local moment description is inadequate.

More recently, a generalized Rhodes–Wohlfarth plot obtained on the basis of the SCR theory of spin fluctuations has been proposed by Takahashi. In this theory, the ratio  $T_C/T_0$  is an important parameter as it characterizes the degree of localization or itineracy of the spin moment. At  $T_C/T_0 \ll 1$ , magnetic materials have a strong itinerant character, while at  $T_C/T_0 \sim 1$ , they exhibit local moment magnetism.<sup>27</sup> Taking the availability of  $M^4-H/M$  in this compound into account, we use the modified Rhodes–Wohlfarth plot to determine the degree of localization or itineracy of the spin moment in  $\text{Fe}_3\text{GeTe}_2$ . The estimated  $T_C/T_0 = 0.096$  ( $T_0 = 2298.7$  K is used here) for  $\text{Fe}_3\text{GeTe}_2$ , which is comparable to those reported for weak ferromagnets, such as  $\text{MnSi}$  (0.13),  $\text{Ni}_3\text{Al}$  (0.0116), and  $\text{ZrZn}_2$  (0.053). When we plot  $P_{\text{eff}}/P_s$  and  $T_C/T_0$  in the so-called Deguchi–Takahashi plot shown in Fig. 12(a),<sup>15,24,28–35</sup> the



**Fig. 12.** (Color online) (a) Generalized Rhodes–Wohlfarth plot and (b) the corresponding log–log plot. The data for  $\text{Fe}_3\text{GeTe}_2$  is plotted as a closed circle. The other plotted data are cited from Refs. 16 and 24, 28–35.

$P_{\text{eff}}/P_s$  for  $\text{Fe}_3\text{GeTe}_2$  is slightly smaller than the generalized Rhodes–Wohlfarth value  $P_{\text{eff}}/P_s \sim 1.4(T_C/T_0)^{-2/3}$ , as can be clearly seen in Fig. 12(b) in the log–log plot. This may be ascribed to the two-dimensionality of  $\text{Fe}_3\text{GeTe}_2$ , as has been pointed by Takahashi, where a smaller  $P_{\text{eff}}/P_s$  will be obtained with decreasing dimensionality from 3D to 2D for the same  $T_C/T_0$ .<sup>2)</sup> Thus, we conclude that  $\text{Fe}_3\text{GeTe}_2$  should be a quasi-2D itinerant ferromagnet.

## 5. Conclusions

We have successfully grown a single crystal of  $\text{Fe}_3\text{GeTe}_2$  by the chemical transport method. The magnetic susceptibility, electrical resistivity, and heat capacity measurements of the crystal confirm the ferromagnetic transition at 220 K. The temperature dependence of magnetic susceptibility obeys the modified Curie–Weiss law quite well in the high-temperature range. The magnetization of  $\text{Fe}_3\text{GeTe}_2$  by applying a magnetic field to the  $c$ -axis was found to show a convex curvature when plotted in an Arrott plot. The linear relation is obeyed in the  $M^4$  vs  $H/M$  plot near  $T_C$ . By adopting the Takahashi theory, we estimated the spin fluctuation parameters  $T_A$  and  $T_0$  to be about 795.8 and 2298.7 K, respectively, which can reproduce the inverse susceptibility  $\chi^{-1}$  quite well. Furthermore, our results indicate that  $\text{Fe}_3\text{GeTe}_2$  is a quasi-2D itinerant ferromagnet.

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