

## Low-temperature magnetic properties of Heusler compounds $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ ( $x=0.1, 0.3, \text{ and } 0.5$ )

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We carried out magnetization  $M(T)$  and specific-heat  $C_p(T)$  measurements of the new Heusler compounds  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  ( $x=0.1, 0.3, \text{ and } 0.5$ ), which exhibit spin-glass freezing.  $M(T)$  has peak- and irreversibility-type anomalies. The temperatures at which these anomalies appear have magnetic field dependence described by the Gabay-Toulouse and de Almeida-Thouless lines in the low-field range. For the magnetic specific heat  $C_m(T)$ , we did not observe a discontinuity indicating long-range magnetic phase transition but a broad hump characteristic of spin-glass freezing.  $C_m(T)$  in the low-temperature range is described as a combination of linear- and quadratic- $T$  terms. The quadratic- $T$  dependence of  $C_m(T)$  is probably associated with excitation of the Ising component of the freezing spins.

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### I. INTRODUCTION

Recently, so-called full-Heusler compounds with the general formula  $X_2YZ$  ( $X$  and  $Y$  are transition elements, and  $Z$  is an  $sp$  element) have attracted much attention in experimental and theoretical studies because some have half metallicity and thus, have potential technological applications. Half metals have a band gap at the Fermi level  $E_F$  for one spin direction in the density of states. Ishida *et al.*<sup>1,2</sup> predicted the existence of half metallicity in the full-Heusler compound  $\text{Co}_2\text{MnZ}$  ( $Z$ : Si or Ge) through a first-principle band calculation. The itinerant electron is 100% spin polarized in ideal half-metallic materials. In experiments, however, the spin polarization is reduced by disorders and defects in the crystalline structure.<sup>3-5</sup> In a theoretical study, it was pointed out that  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  is a strong candidate material in which the half metallicity is preserved in the presence of higher level defects and randomness of the crystalline structure.<sup>6</sup> Hiroi *et al.*<sup>7-9</sup> synthesized  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  compounds over a wide range of  $x$ , and investigated their transport and magnetic properties. For  $1.4 \leq x < 2.0$ ,  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  shows ferromagnetic transition at a Curie temperature  $T_C \sim 500$  K. With  $x$  decreasing from  $x \sim 1.4$ ,  $T_C$  decreases linearly, and the system becomes antiferromagnetic below  $x=0.2$ . In addition, for  $x \leq 0.5$ , irreversibility between the magnetization  $M(T)$  of zero-field cooling (ZFC) and field cooling (FC) is observed. This behavior is strongly suggestive of spin-glass freezing. Meanwhile, the metallic transport property in the higher  $x$  range changes to an insulating property around  $x=0.7$  with decreasing  $x$ .

In this paper, we report the magnetization and specific heat of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , especially for samples within the insulating region,  $x \leq 0.5$ . We suggest that the quadratic temperature dependence of magnetic specific heat in this system is caused by excitation of the Ising component of the freezing spins.

### II. EXPERIMENTAL

The polycrystalline samples of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  ( $x=0.1, 0.3, 0.5, \text{ and } 1.6$ ) were prepared employing the arc-melting method with high-purity constituent elements under an argon atmosphere. All samples were confirmed by x-ray powder diffraction to be of single phase and have the  $L2_1$ -type crystal structure shown in Fig. 1. The lattice constant  $a$  decreases monotonically with increasing  $x$ . For  $x=0.1, 0.3, 0.5, \text{ and } 1.6$ ,  $a=5.88, 5.87, 5.85, \text{ and } 5.71$  Å are obtained, respectively. In  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , the effect of the substitution of Ru for Fe atoms on the atomic order in the  $L2_1$  structure is unclear. Recently, neutron-diffraction studies on analog compounds  $\text{Ru}_{2-x}\text{Fe}_x\text{CrGe}$  were carried out by Brown *et al.*<sup>10</sup> They reported that for  $x=0$ , Ru, Cr, and Ge atoms are highly ordered at the X, Y, and Z sites, respectively; however, substitution of Ru for Fe atoms leads to disorder of the Cr and Fe among the X and Y sites. Also in  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , atomic

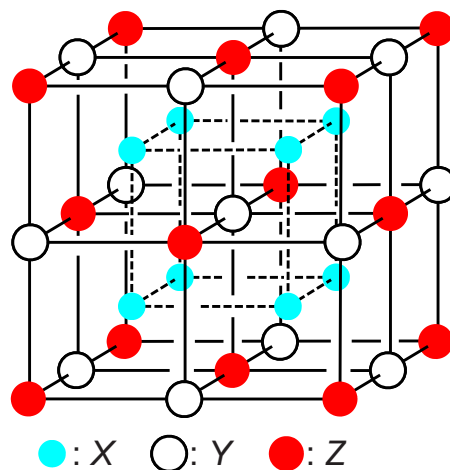


FIG. 1. (Color online)  $L2_1$  structure of the Heusler compounds  $X_2YZ$ . The space group is  $Fm\bar{3}m$ .

disorder similar to that of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrGe}$  might occur owing to element substitution.

The dc magnetization  $M(T)$  measurements were carried out using a superconducting quantum interference device magnetometer (Quantum Design MPMS). Specific heat  $C_p$  in the temperature range between 1.4 and 300 K was measured using a home-built automated calorimeter, which is operated employing the adiabatic heat-pulse method. The performance of the calorimeter was tested by comparing the specific heat of copper reported previously,  $C_{ref}$ ,<sup>11,12</sup> with our result  $C_{exp}$  measured using 0.266 g of copper. The deviation from the reference value ( $=|(C_{ref}-C_{exp})/C_{ref}|$ ) was within 2% above 100 K and within 5% below 100 K. Samples of 1.700, 2.570, 3.106, and 0.787 g mass were used to measure  $C_p$  in the cases of  $x=0.1, 0.3, 0.5$ , and 1.6 for  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , respectively.

### III. RESULTS AND DISCUSSION

#### A. Spin-glass behavior at low temperature

Figures 2(a)–2(c) show the temperature  $T$  dependence of the dc magnetic susceptibility, which is obtained by dividing  $M(T)$  by the magnetic field  $H=1$  kOe,  $M(T)/H$ , for  $x=0.1, 0.3$ , and  $0.5$ , respectively. For  $x=0.1$ ,  $M(T)/H$  shows a cusplike peak anomaly at  $T_{f1}=33$  K, and strong irreversibility between  $M(T)/H$  values of FC and ZFC appears below  $T_{f2}=15$  K.  $M(T)/H$  of  $x=0.3$  also shows peak- and irreversibility-type anomalies in the low-temperature range. For  $x=0.5$ , these anomalies remain, although the peak-type anomaly at  $T_{f1}$  is broad. Inverted magnetic susceptibility,  $[M(T)/H]^{-1}$ , is plotted in the inset of each panel of Fig. 2. For  $x=0.1$ , from the fitting with the Curie-Weiss law,  $C/(T-\theta)$ , to the data above 200 K, the Weiss temperature  $\theta=-111$  K and the Curie constant  $C=0.774$  emu K/mol are obtained. The negative value of  $\theta$  means that the magnetic interaction in the system is mainly antiferromagnetic. From the value of  $C$ , the effective magnetic moment  $\mu_{eff}=2.49 \mu_B$  is obtained. By increasing  $x$  to 0.3, the value of  $\theta$  becomes positive ( $\theta=164$  K), meaning that the ferromagnetic interaction overcomes the antiferromagnetic one. The estimated value of  $\mu_{eff}$  from  $C$  is  $2.59 \mu_B$ . For  $x=0.5$ , the value of  $\theta$  increases to 216 K; i.e., increasing  $x$  enhances the ferromagnetic nature of the system. The value of  $\mu_{eff}$  ( $=3.25 \mu_B$ ) is larger than that for  $x=0.1$  and 0.3. Unfortunately, in this system, it is unclear which atom is responsible for the magnetic moment at this stage.<sup>7,8</sup>

Figures 3(a)–3(c) are plots of specific heat  $C_p(T)$  versus  $T$  in the temperature range between 1.4 and 80 K for  $x=0.1, 0.3$ , and  $0.5$ , respectively. No discontinuity indicating long-range order is observed for any of the samples around the temperature at which  $M(T)/H$  has peak-type (at  $T_{f1}$ ) and irreversibility-type (at  $T_{f2}$ ) anomalies. As is well known,  $C_p(T)$  of the spin-glass system does not have discontinuity.<sup>13–15</sup> Judging from the magnetic and thermodynamic properties, the anomalies of  $M(T)/H$  at  $T_{f1}$  and  $T_{f2}$  may be caused by the spin-glass freezing.

#### B. Magnetic field dependence of the spin-glass freezing temperature

To investigate the anomalies of  $M(T)$  in detail, we measured ZFC and FC  $M(T)$  for the various magnetic fields  $H$ .

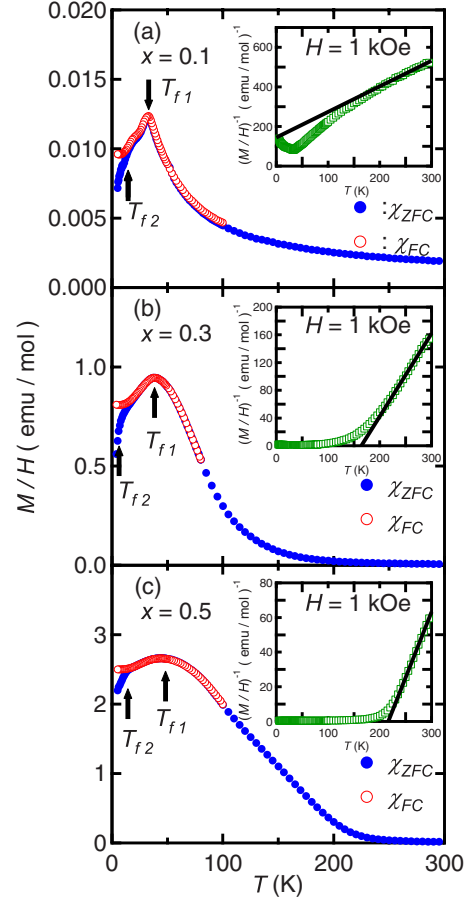


FIG. 2. (Color online) Temperature  $T$  dependence of dc magnetic susceptibility  $M(T)/H$  in the range  $5 \leq T \leq 300$  K for  $\text{Ru}_{2-x}\text{Fe}_x\text{Si}$  with (a)  $x=0.1$ , (b)  $x=0.3$ , and (c)  $x=0.5$ . Measurements were carried out with the magnetic field  $H=1$  kOe. The arrows show the position of the peak-type ( $T_{f1}$ ) and irreversibility-type ( $T_{f2}$ ) anomalies. The inset shows the inverted susceptibility  $[M(T)/H]^{-1}$  in the range  $5 \leq T \leq 300$  K. The solid line is a best fit of the Curie-Weiss law.

Figures 4(a)–4(c) show the  $T$  dependence of  $M(T)$  for representative  $H$  with  $x=0.1, 0.3$ , and  $0.5$ , respectively.

As shown by the arrows, we have determined  $T_{f1}$  and  $T_{f2}$  as the temperatures at which  $M(T)$  shows a peak and a kink in the ZFC process, respectively. For all  $x$  concentrations, with increasing  $H$ ,  $T_{f2}$  decreases rapidly. The  $H$  dependence of  $T_{f1}$  and  $T_{f2}$  for  $x=0.1, 0.3$ , and  $0.5$  is summarized in Figs. 5(a)–5(c), respectively.

From a theoretical perspective, de Almeida and Thouless studied the Ising-spin-glass system on the basis of the Sherrington-Kirkpatrick (SK) model.<sup>16</sup> They predicted that the spin freezing temperature  $T_{AT}$  depends on  $H$ . In the low  $H$  range,  $T_{AT}$  varies as shown in the so-called de Almeida-Thouless (AT) line,

$$H_{AT}(T_{AT}) = A_{AT} \left[ 1 - \frac{T_{AT}(H)}{T_{AT}(0)} \right]^{3/2}. \quad (1)$$

The AT line has been observed for materials with Ising spins in experimental studies.<sup>17,18</sup> In addition, Gabay and

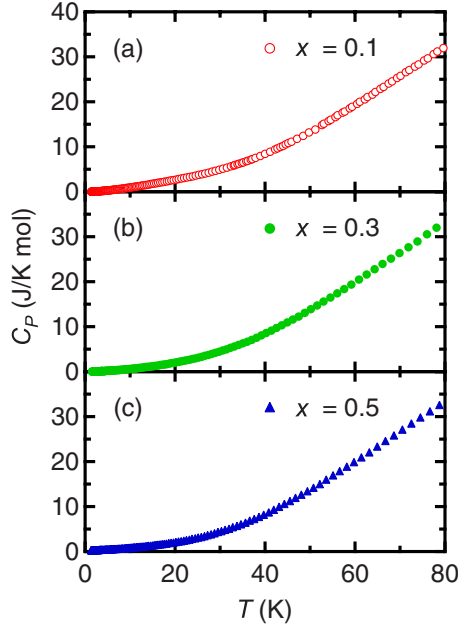


FIG. 3. (Color online) Temperature dependence of the specific heat  $C_p(T)$  in the range  $1.4 \leq T \leq 80$  K for  $\text{Ru}_{2-x}\text{Fe}_x\text{Si}$  with (a)  $x = 0.1$ , (b)  $x = 0.3$ , and (c)  $x = 0.5$ .

Toulouse<sup>19</sup> investigated the  $H$  dependence of the spin freezing temperature for the Heisenberg spin-glass system. In this model, in addition to  $T_{\text{AT}}$ , another spin freezing temperature,  $T_{\text{GT}} (> T_{\text{AT}})$ , appears by applying  $H$ . Namely, successive spin-glass freezing occurs. The  $H$  dependence of  $T_{\text{GT}}$  is called the Gabay-Toulouse (GT) line, expressed as

$$H_{\text{GT}}(T_{\text{GT}}) = A_{\text{GT}} \left[ 1 - \frac{T_{\text{GT}}(H)}{T_{\text{GT}}(0)} \right]^{1/2}. \quad (2)$$

In the  $T$  range between  $T_{\text{AT}}$  and  $T_{\text{GT}}$ , the transverse component of the spin moments freezes with the remaining degrees of freedom of the longitudinal component. Below  $T_{\text{AT}}$ , the longitudinal component freezes and the spin moments completely freeze. The existence of successive spin-glass freezing and the  $H$  dependence of these two lines have been confirmed in experimental studies.<sup>20,21</sup>

The solid and dashed lines in Figs. 5(a)–5(c) represent the results of fitting with the AT [Eq. (1)] and GT lines [Eq. (2)], respectively. For all values of  $x$ , the  $H$  dependence of  $T_{f2}$  agrees with the AT line in the low  $H$  range. This suggests that the large irreversibility of  $M(H)$  at  $T_{f2}$  results from AT freezing; namely, the longitudinal component freezing. In the higher  $H$  range, our results deviate from the AT line. This is because Eq. (1) is an approximate expression for the low  $H$  range.<sup>16,18</sup> The value of  $T_{f2}$  at  $H = 0$  kOe,  $T_{f2}(0)$ , seems to be independent of  $x$ ;  $T_{f2}(0) \sim 20$  K. As shown in Figs. 5(a) and 5(b), for  $x = 0.1$  and 0.3, the  $H$  dependence of  $T_{f1}$  obeys the GT line, and suggests that GT freezing (the transverse component freezing) occurs at  $T_{f1}$ . Meanwhile, for  $x = 0.5$ , the  $H$  dependence of  $T_{f1}$  is not reproduced by the GT line. However, from analogy with the cases of  $x = 0.1$  and 0.3, we consider that also for  $x = 0.5$ , the origin of the peak-type anomaly of  $M(H)$  at  $T_{f1}$  is the occurrence of GT freezing. Deviation

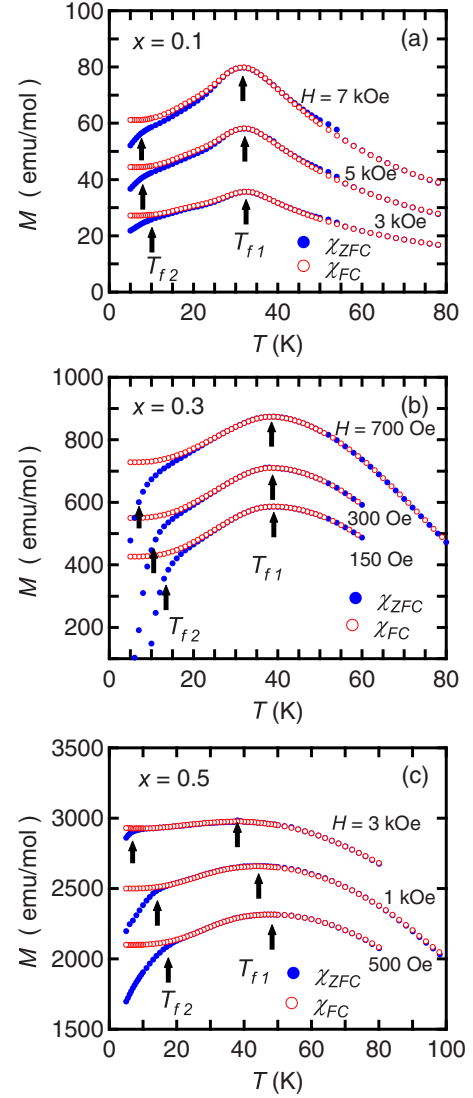


FIG. 4. (Color online) Temperature dependence of magnetization  $M(T)$  in representative magnetic field  $H$  for  $\text{Ru}_{2-x}\text{Fe}_x\text{Si}$  with (a)  $x = 0.1$ , (b)  $x = 0.3$ , and (c)  $x = 0.5$ . The arrows show the position of peak-type ( $T_{f1}$ ) and irreversibility-type ( $T_{f2}$ ) anomalies.

from the theoretical GT line might result from the difficulty of determining the peak position of  $M(T)$  because of the peak broadening due to the large ferromagneticlike moment.

In general, the AT and GT lines merge at  $H = 0$  kOe. It is interesting that the AT line separates from the GT line at  $H = 0$  kOe in our system. The separation of the two lines in the absence of a magnetic field might be assisted by the ferromagnetic internal field, which probably has a short range. A similar behavior has been reported for the compound  $\text{Ni}_{177}\text{Mn}_{23}$ .<sup>22</sup>

### C. Magnetic excitation at low temperature

As mentioned in Sec. I, the electrical transport property of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  depends on  $x$ . The property is insulating for  $x \leq 0.7$ , whereas it is metallic for  $x > 0.7$ . Figure 6 shows the specific heat divided by temperature  $C_p/T$  versus  $T$  in the range  $1.4 \leq T \leq 300$  K for the insulator  $x = 0.1, 0.3$ , and 0.5,

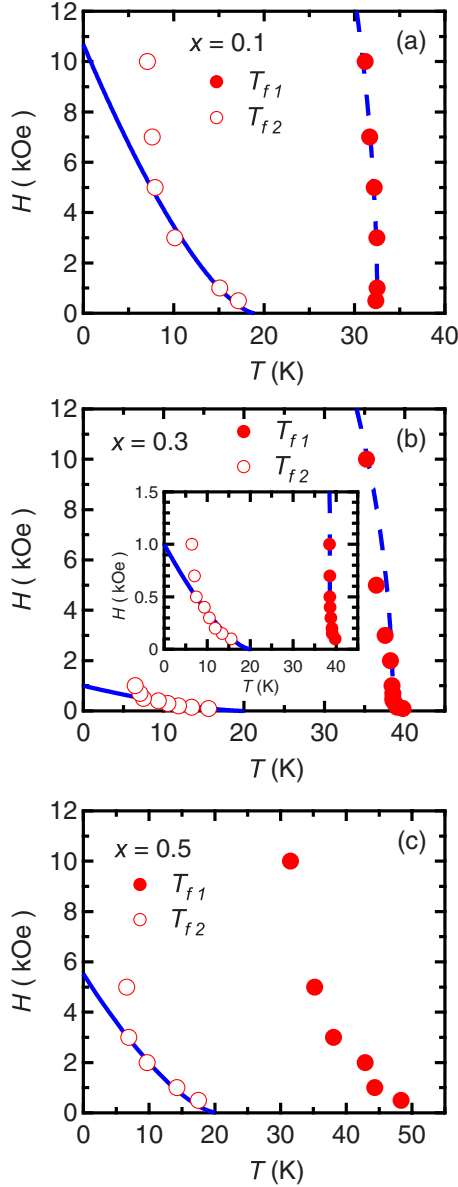


FIG. 5. (Color online) Magnetic field dependence of  $T_{f1}$  and  $T_{f2}$  for  $\text{Ru}_{2-x}\text{Fe}_x\text{Si}$  with (a)  $x=0.1$ , (b)  $x=0.3$ , and (c)  $x=0.5$ . The solid and dashed lines are fits with the AT line [Eq. (1)] and GT line [Eq. (2)], respectively. The inset of (b) is the plot with an expanded vertical scale.

with data for the metal corresponding to  $x=1.6$  shown for comparison. The discontinuity of specific heat as seen in the first- or second-order phase transitions is not observed even in the wide temperature range. For the metallic specimen corresponding to  $x=1.6$ , as shown by the solid line in the inset of Fig. 6, which is a plot of  $C_p(T)/T$  as a function of  $T^2$ , the low-temperature part of the specific-heat curve can be fitted by Debye's law,  $C_p(T) = \gamma T + \beta T^3$ , with  $\gamma = 2.37 \times 10^{-2} \text{ J/K}^2 \text{ mol}$  and  $\beta = 6.38 \times 10^{-5} \text{ J/K}^4 \text{ mol}$ . Here, the first and second terms are the conduction electron and lattice contributions, respectively. Using the value of  $\beta$ , the Debye temperature  $\theta_D = 496 \text{ K}$  is obtained from the relation  $\beta = 12\pi^4 n N_A k_B / 5 \theta_D^3$ , where  $n$  is the number of atoms ( $n=4$  in

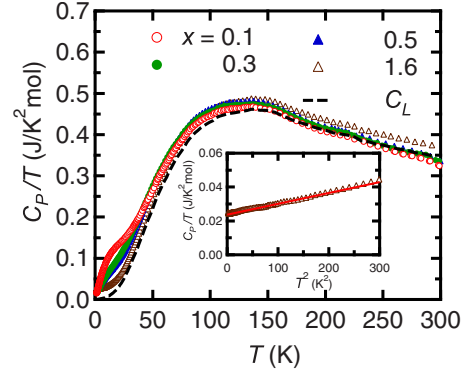


FIG. 6. (Color online) Temperature dependence of specific heat divided by temperature,  $C_p(T)/T$ , of  $\text{Ru}_{2-x}\text{Fe}_x\text{Si}$  for  $x=0.1, 0.3, 0.5$ , and  $1.6$ . The dashed line is the lattice contribution  $C_L(T)$  estimated from  $C_p(T)$  for  $x=1.6$ . The inset shows  $C_p(T)/T$  versus  $T^2$  for  $x=1.6$ . The solid line shows Debye's  $T^3$  law.

this system),  $N_A$  is the Avogadro's number, and  $k_B$  is the Boltzmann's constant.

To obtain the temperature dependence of the magnetic specific heat  $C_m(T)$  for insulating  $x=0.1, 0.3$ , and  $0.5$ , we tried to estimate the lattice specific heat  $C_L(T)$  of this system by subtracting the conduction-electron part  $C_e(T) (= \gamma T)$  from  $C_p(T)$  for  $x=1.6$ . The obtained  $C_L(T)$  is shown as the dashed line in Fig. 6. The  $x=1.6$  specimen is the itinerant-magnetic system and has a ferromagnetic phase transition at a higher temperature,  $T_c \sim 500 \text{ K}$ ,<sup>7</sup> as mentioned in Sec. I. However, we suppose that the magnon contribution to  $C_p(T)$  for  $T < 300 \text{ K}$  is negligible. Since  $C_p(T)$  for  $x=0.1, 0.3$ , and  $0.5$  converge to the estimated  $C_L(T)$  in the higher temperature range, as shown in Fig. 6, this assumption may be reasonable. Figure 7(a) shows  $C_m(T)$  versus  $T$  obtained from  $C_p(T) - C_L(T)$  for  $x=0.1, 0.3$ , and  $0.5$  in the range  $1.4 \leq T \leq 120 \text{ K}$ . For all samples,  $C_m(T)$  has a broad hump. The appearance of the broad hump in the specific heat is a strong characteristic of the spin-glass system.<sup>14</sup> The peak position of  $C_m(T)$  shifts to higher temperature with increasing  $x$ .

The magnetic specific heat divided by temperature  $C_m(T)/T$  versus  $T$  is shown in Fig. 7(b). The temperature at the maximum  $C_m(T)/T$ ,  $T_{\max}$ , almost coincides with the AT-freezing temperature at  $H=0 \text{ kOe}$  determined from  $M(T)$ ,  $T_{f2}(0) \sim 20 \text{ K}$  for all samples. This means that the spin components frozen at  $T_{f2}(0)$  greatly contribute to the  $T$  dependence of  $C_m(T)$  in the low  $T$  ( $\leq T_{f2}$ ) range because  $C_m(T)/T$  is equivalent to the temperature derivative of magnetic entropy,  $dS_m/dT$ .

The low-temperature part of the  $C_m(T)/T$  curve is shown in Fig. 8 and  $C_m(T)$  can be described as the simple combination of linear and quadratic terms,  $C_m(T) = \alpha_1 T + \alpha_2 T^2$ , as shown by the solid lines. For  $x=0.1$ ,  $\alpha_1$  is almost zero, whereas for  $x=0.3$  and  $0.5$ ,  $\alpha_1$  has a finite value. This  $T$  dependence is in contrast to the simple linear- $T$  dependence of  $C_m(T)$  for the low-temperature excitation of conventional spin-glass materials.<sup>13</sup> We will discuss the  $T$  dependence of  $C_m(T)$  later.

Magnetic entropy  $S_m(T)$  is derived by integrating  $C_m(T)/T$  with respect to  $T$ , and is shown in Fig. 9. In a previous investigation, from the analysis of the Curie constant, spin

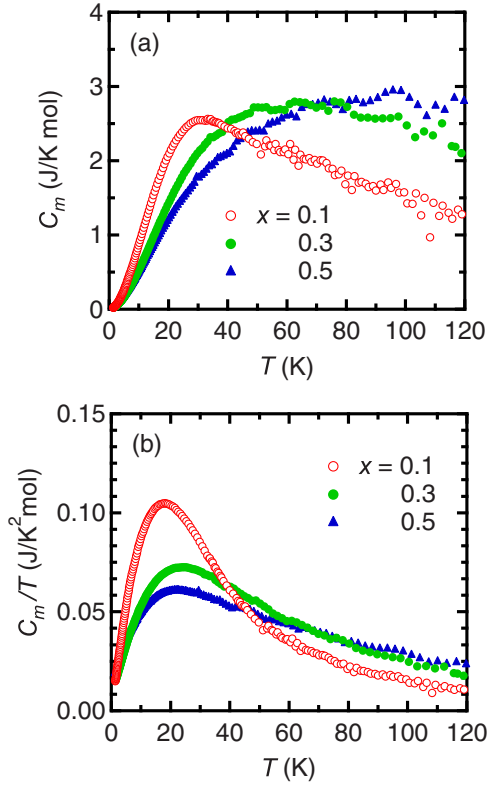


FIG. 7. (Color online) (a) Temperature dependence of the magnetic specific heat  $C_m(T)$  of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  for  $x=0.1, 0.3$ , and  $0.5$  in the range  $1.4 \leq T \leq 120$  K. (b) Temperature dependence of magnetic specific heat divided by temperature  $C_m(T)/T$ .

angular momentum  $S=1$  was predicted for these concentrations.<sup>8</sup> This  $S$  value leads to  $S_m = R \ln(3) = 9.13$  J/K mol. Here,  $R$  is the gas constant. However, the value of  $S_m$  obtained from specific heat is much smaller than  $R \ln(3)$  for all concentrations, even at  $T=120$  K. This might imply that the amount of localized spin contributing to spin-glass freezing is small, or a finite degree of freedom remains for  $T < 1.4$  K.

We now discuss the quadratic- $T$  dependence of  $C_m(T)$  in the low  $T$  range for our system. As mentioned above, this is in contrast to the linear- $T$  dependence seen in the conven-

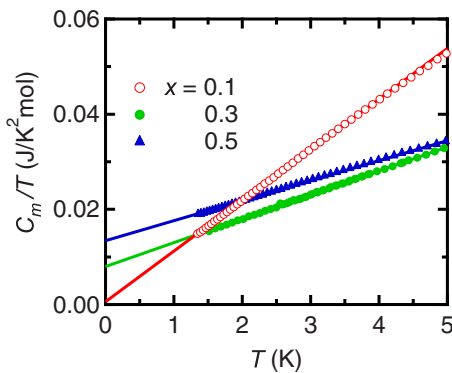


FIG. 8. (Color online) Temperature dependence of  $C_m/T$  of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  for  $x=0.1, 0.3$ , and  $0.5$  in the range  $1.4 \leq T \leq 5$  K. The solid lines are linear fits.

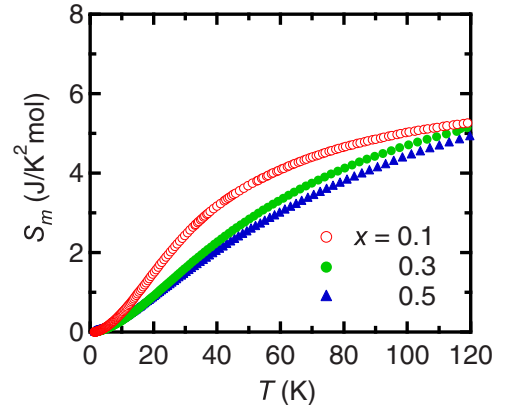


FIG. 9. (Color online) Temperature dependence of magnetic entropy  $S_m(T)$  of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  for  $x=0.1, 0.3$ , and  $0.5$  in the range  $1.4 \leq T \leq 120$  K.

tional spin-glass system. Thouless, Anderson, and Palmer<sup>23</sup> (TAP) investigated the solution of the SK model mentioned in Sec. III B without using a replica method. This model has a Hamiltonian with exchange interaction  $J_{ij}$  between the nearest-neighbor Ising spins  $S_i$  and  $S_j$ ,

$$H = - \sum_{(ij)} J_{ij} S_i \cdot S_j, \quad (3)$$

where the sum is over the nearest-neighbor pairs. The value of  $J_{ij}$  is independently distributed with a Gaussian probability distribution,

$$P(J_{ij}) \propto \exp\left(\frac{-NJ_{ij}^2}{2\tilde{J}^2}\right) \quad (4)$$

with a variance  $\tilde{J}^2/N$  and where  $N$  is the number of neighbors of each spin. They pointed out that  $C_m(T)$  for this model shows quadratic- $T$  dependence,

$$C_m(T) \propto N \left(\frac{T}{\tilde{J}}\right)^2, \quad (5)$$

below the spin-glass freezing temperature. This freezing temperature coincides with the AT line for zero magnetic field. As mentioned in Sec. III B, for  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , the AT line is separated from the GT line even for a zero magnetic field. Therefore, the magnetic excitation in the  $T$  range below the AT line is mainly the contribution from the Ising component of the freezing spins; i.e., the system can be regarded as an ideal Ising-spin-glass system well described by TAP and AT theories. This situation may give rise to the quadratic- $T$  dependence of  $C_m(T)$  as expressed by Eq. (5).

To compare with each  $T^2$  dependence of  $C_m(T)$  for  $x=0.1, 0.3$ , and  $0.5$ , we determined  $C_m^*(T) = C_m(T) - \alpha_1 T$ . Figure 10(a) shows plots of  $C_m^*/T$  divided by its maximum value  $(C_m^*/T)_{T=T_{\max}}$  versus the reduced temperature  $T/T_{\max}$ . It is noteworthy that the curves for  $(C_m^*/T)/(C_m^*/T)_{T=T_{\max}}$  converge below  $T/T_{\max}=1$  for all  $x$  concentrations, although the mainly magnetic interaction ( $J_{ij} \sim \theta$ ), and even its sign, strongly depends on  $x$ , as mentioned in Sec. III A. As shown in Fig. 10(b), the coefficient of  $T^2$  is almost the same for all

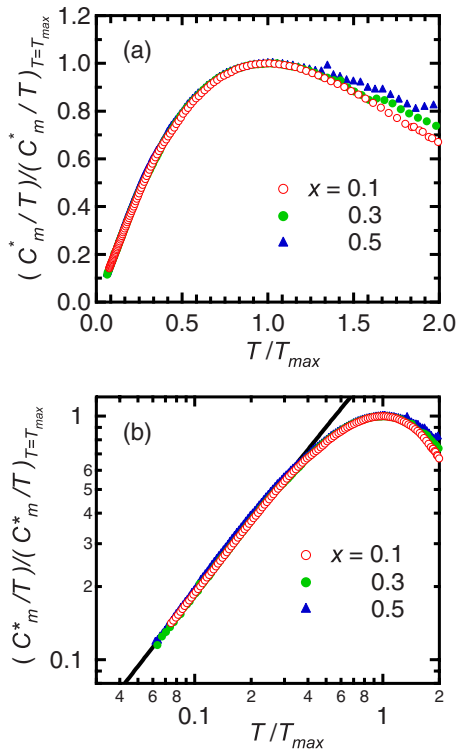


FIG. 10. (Color online) Plots of  $C_m^*/T$  normalized by its maximum value  $(C_m^*/T)_{T=T_{\max}}$ ,  $(C_m^*/T)/(C_m^*/T)_{T=T_{\max}}$ , versus reduced temperature  $T/T_{\max}$  of  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  for  $x=0.1, 0.3$ , and  $0.5$  with a (a) linear and (b) logarithmic scale. The solid line in (b) shows  $T^2$  dependence.

$x$ . This might suggest that  $\tilde{J}$  is insensitive to  $x$  in contrast to  $J_{ij}$ .

Finally, we mention the linear temperature dependence of  $C_m(T)$  in this system. Presently, there is no firm understand-

ing of the origin of the linear dependence. The amplitude of linear- $T$  dependence,  $\alpha_1$ , increases with increasing  $x$ , as shown in Fig. 8. It is well known that  $C_m(T)$  of the conventional spin-glass system has linear- $T$  dependence, as mentioned above. The linear- $T$  dependence of  $C_m(T)$  in the present system might come from the excitation of the transverse component of the freezing spins. Another possibility is the existence of the residual density of states. In  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$ , the transport property in the Fe-rich range ( $x > 0.7$ ) is metallic. On the other hand, the samples with  $x \leq 0.5$  and spin-glass freezing are insulators. If residual density of states exists at the Fermi level for the insulator samples,  $C_m(T)$  has the linear- $T$  term. Detailed studies are needed to clarify the unusual low-temperature magnetic excitation in this system.

#### IV. CONCLUSION

We studied the magnetic and thermodynamic properties of the new Heusler compounds  $\text{Ru}_{2-x}\text{Fe}_x\text{CrSi}$  with  $x=0.1, 0.3$ , and  $0.5$ . All samples show peak- and irreversibility-type anomalies in magnetization  $M(T)$ . Because no discontinuity indicating long-range magnetic order is observed in the specific heat  $C_p(T)$  of each sample, spin-glass freezing occurs at around the temperatures at which the anomalies appear in  $M(T)$ . In a weak magnetic field, the peak- and irreversibility-type anomalies of  $M(T)$  depend on the magnetic field  $H$  and can be described by the GT and AT lines, respectively. Even at zero magnetic field, the AT line is separated from the GT line for all samples. Quadratic temperature dependence of magnetic specific heat  $C_m(T)$  is observed in the low-temperature range. We suggest that this might be due to the excitation of the Ising component of the freezing spins. Magnetic specific heat divided by temperature  $C_m(T)/T$  has a broad hump around the AT-freezing point for zero magnetic field.

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