Ferromagnetism and spin-glass transitions in the Heusler compounds $Ru_{2-x}Fe_xCrSi$

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The results of a comprehensive study of the structural, magnetic, and magnetotransport properties of the Heusler compounds $Ru_{2-x}Fe_xCrSi$ are presented. The Fe-rich compounds ($x \geq 1.5$) exhibit a usual ferromagnetic transition. The Ru-rich compound ($x=0.1$) does not show ferromagnetism but exhibits a peak in magnetic susceptibility at $T_N^*=30$ K. Nevertheless, specific-heat measurements show that there is no antiferromagnetic transition at T_N^* nor at any other temperatures. With further decreasing temperature strong irreversible behavior occurs below a temperature T_g. It is proposed that these results can be interpreted as successive spin-glass transitions at T_N^* and T_g. The compounds with intermediate x (=0.3 and 0.5) are found to be ferromagnetic. Both the saturation magnetization and the Curie temperature T_C increase with increasing x. However, being different from the Fe-rich compounds, hysteresis of the magnetization is observed and this suggests that the ferromagnetism has glassy character. In lower-temperature range fairly large negative magnetoresistance are observed. These results are suggestive of a magnetically inhomogeneous ferromagnetic state, that is, the formation of ferromagnetic clusters.

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I. INTRODUCTION

Heusler compounds have been studied for a long time mainly because of their unique magnetism. Heusler compounds have the formula X_2YZ, where X and Y are transition metals and Z are sp elements. They crystallize in cubic $L2_1$-type structure which consists of four penetrating fcc sublattices (Fig. 1). Recently they have been attracting growing interests because of their potential use as half metals (HMs),[1,2] ferromagnetic-shape memory alloys, and thermoelectric materials. Particularly, HMs are expected to be utilized in so-called *spintronics*, for example, tunneling magnetoresistance devices.

A HM possesses states at the Fermi energy only in one spin band while a gap in the other spin band and thus it is a ferromagnetic metal with its conduction electrons 100% spin polarized. The concept of the half metal is first introduced in NiMnSb, which belongs to $C1_b$ structure compounds called half-Heusler compounds.[3] In full-Heusler compounds with $L2_1$ structure, which is often just called Heusler compounds, Ishida *et al.* predicted that the Heusler compound Co_2MnZ, where Z stands for Si or Ge, is a HM from first-principle band-structure calculations.[4,5] Since then HMs have been predicted also in other Heusler and half-Heusler compounds.[1,2,6] Experimentally, however, the realization of half metallicity in Heusler compounds was not sufficiently successful. It was revealed both theoretically and experimentally that disorders and surfaces depress the half metallicity.[5,7–9] However, it was only recently that, by fabricating a magnetic tunneling junction consisting of Co_2MnSi, the observation of a magnetoresistance ratio of 159% at 2 K was achieved.[10] This result demonstrated that Co_2MnSi is intrinsically a HM. Although it is interesting to find idealistic HMs, ferromagnetic metals with high spin polarization which are insensitive to disorders, even if they are not complete HMs, are highly valuable particularly in light of application. A Heusler-type compound $Co_2Cr_{0.6}Fe_{0.4}Al$ was sug-

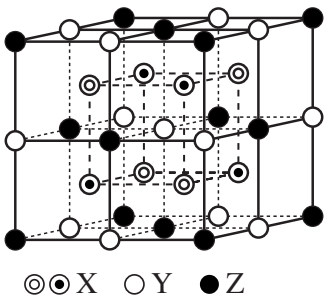

FIG. 1. $L2_1$ crystal structure of the Heusler compounds X_2YZ.

gested to be such a material.[11,12] In this material the high spin polarization should be preserved even when the minority-spin states appear by disorders, because the density of states of the majority-spin states is supposed to be large at the Fermi energy.[13] From a similar viewpoint Heusler compounds $Ru_{2-x}Fe_xCrSi$ were predicted to be complete or nearly complete HMs which are robust to chemical disorders by a first-principle band-structure calculation.[14,15]

Motivated by the theoretical results we prepared a series of the Heusler compounds $Ru_{2-x}Fe_xCrSi$.[16,17] We previously reported that $Ru_{2-x}Fe_xCrSi$ is ferromagnetic for $x \geq 0.5$, and for $x \gtrsim 1.5$ its Curie temperature T_C is much higher than room temperature.[16] The saturation magnetization extrapolated to 0 K M_0 for $x \sim 2$ is nearly $2\mu_B$, which is close to the theoretical prediction.[14] From these results $Ru_{2-x}Fe_xCrSi$ turned out to be a promising candidate for a material with high spin polarization which is robust against disorders and to be potentially useful in the area of spintronics.

There is discrepancy between the theory and the experiments in these series. The theory shows that the value of M_0 is $\sim 2\mu_B$ and independent of x, while we found that M_0 obtained from magnetization measurements decreases with decreasing x. Further we found that T_C decreases with decreasing x, and for $x=0.1$ ferromagnetism disappears and a cusp-shape anomaly appears at 30 K in magnetic susceptibility which is likely to indicate an antiferromagnetic transition.[17] At lower temperatures the separation of magnetic susceptibility between under zero-field-cooling condition (ZFC) and field-cooling condition (FC), which indicates a spin-glass (SG) state was found.

For the Ru-rich compounds the band-structure calculation demonstrated another interesting result. The theory revealed that for $x \leq 0.5$ antiferromagnetic states are energetically preferable to the ferromagnetic state,[14] while the experimental results indicate that competition between ferromagnetism and antiferromagnetism is present, and that this competition yields complex magnetic states, not a simple antiferromagnetic state. As Heusler compounds have often been considered to be typical local-moment ferromagnets, only a few are known to be antiferromagnets. In antiferromagnetic Heusler compounds frustration may be inherent because in Heusler structure each sublattice forms a fcc structure, and it is accepted that in fcc structures several possible magnetic-ordered states with ferromagnetic and antiferromagnetic components have close energy. Actually in $Ru_{2-x}Fe_xCrSi$ case, it was theoretically shown that possible ferromagnetic and a few antiferromagnetic states are close in energy, and this would cause frustration. The existence of the frustration is consistent with the experimental results that the SG state appears. However, thus far much attention has not been drawn to the frustration in the magnetism in Heusler compounds.

Another interesting finding was a metal-semiconductor crossover induced by varying composition.[17] The Fe-rich compounds exhibit metallic behavior in electrical resistivity, while semiconducting behavior is seen for $x \lesssim 0.7$. The origin of the metal-semiconductor crossover has not been understood yet.

In order to understand this series of compounds $Ru_{2-x}Fe_xCrSi$, it is necessary to investigate their physical prosperities more extensively. In this paper we will present results of a comprehensive study on magnetic and transport properties of a series of the Heusler compounds $Ru_{2-x}Fe_xCrSi$ for a wide range of x, including studies in high magnetic field and under pressure. Particularly SG-like behavior for $x=0.1$ is intensively investigated. Part of the results were reported in Refs. 16 and 17.

II. EXPERIMENTS

Polycrystalline samples were prepared by arc melting high-purity constituent elements under high-purity argon atmosphere. Since the noticeable effects of the annealing of the samples were not observed, we report here on arc-melted samples. Crystal structure was examined by x-ray powder-diffraction measurements with Cu $K\alpha$ radiation, and the temperature dependence of diffraction patterns was measured with a high-temperature attachment and a cryostat.

Magnetization (M) was measured with commercial superconducting quantum interference device magnetometers (MPMS, Quantum Design). To reveal hysteresis behavior, experiments under ZFC and FC were performed. In ZFC, a sample was first cooled to a low temperature (T) (2 K in most cases) from a sufficiently high T under zero magnetic field (H); at this temperature H was applied and then magnetization $M_{\mathrm{ZFC}}(T)$ was measured at this constant H with increasing T. In FC, first H was applied at a sufficiently high T, keeping H constant, the sample was cooled to the lowest T, and then the magnetization $M_{\mathrm{FC}}(T)$ was measured at this H. Specific-heat measurement from 2 to 100 K was carried out by a conventional adiabatic heat-pulse method. In the measurements of M under high pressure we generated hydropressures up to 1.7 GPa with a CuBe and NiCrAl hybrid piston cylinder cell. The measurements of M in high H up to 18 T were performed by an induction magnetometer in a superconducting magnet. Electrical resistivity (ρ) was measured by the usual four-probe method and the measurements of ρ in H up to 7 T were performed with a Quantum Design physical properties measurement system (PPMS).

III. RESULTS

A. Crystal structure

We prepared polycrystalline samples of $Ru_{2-x}Fe_xCrSi$ for a wide range of Fe concentration x ($0.1 \leq x \leq 1.8$). Thus far we have not succeeded in synthesizing single-phase samples of the end materials with $x=0$ and 2. By x-ray diffraction measurements each synthesized sample was confirmed to be single phase. Profiles of x-ray diffraction pattern for $x=0.3$ with Cu $K\alpha$ radiation at various T ($20 \leq T \leq 290$ K) are shown in Fig. 2(a). The patterns show that the crystal structure is $L2_1$. Reflections (111) and (200) are superlattice reflections. Their structure factors are $F(111)=4|f_Y-f_Z|$ and $F(200)=4|2f_X-(f_Y+f_Z)|$, respectively, where f_i ($i=X,Y,Z$) are the average scattering factors for the i sites. They are signs of the atomic order in the $L2_1$ structure. On the other hand, the structure factor of the (220) reflection is given by $F(220)=4|2f_X+f_Y+f_Z|$. This is a principal lattice peak and the intensity is not affected by the atomic disorder. Figure

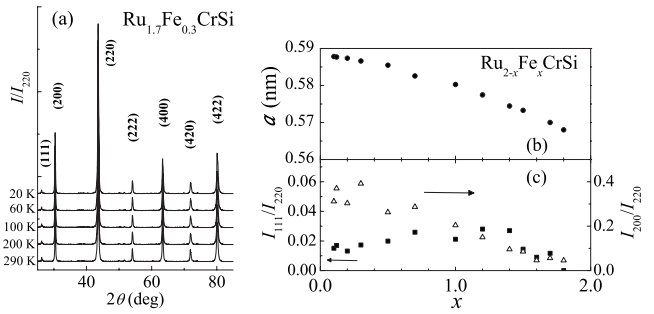

FIG. 2. (a) X-ray diffraction patterns of $Ru_{1.7}Fe_{0.3}CrSi$ with Cu $K\alpha$ radiation at various temperatures from 20 to 290 K. (b) Lattice constant a of $Ru_{2-x}Fe_xCrSi$. (c) Relative diffraction intensities of (111) and (200) superlattice peaks divided by the intensity of the principle peak (220), I_{111}/I_{220} (solid squares) and I_{200}/I_{220} (open triangles), are shown as a function of x. They represent Y-Z and X-YZ atomic orders in the Heusler structure (X_2YZ), respectively.

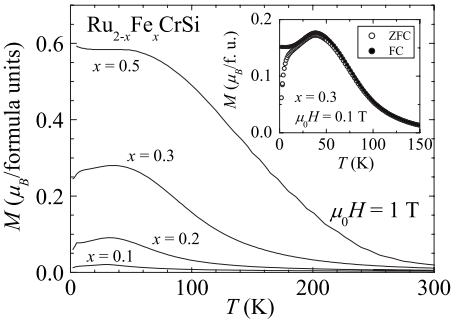

FIG. 3. Temperature dependence of magnetization M of $Ru_{2-x}Fe_xCrSi$ (x=0.1, 0.2, 0.3, and 0.5) in μ_0H=1 T. The data were taken under zero-field-cooling (ZFC) conditions. In the inset magnetization for x=0.3 at a lower magnetic field (0.1 T) is shown. The peak and the hysteresis are pronounced as observed for smaller x in 1 T.

2(b) shows the lattice constant a with various x. Figure 2(c) shows the relative intensity of the x-ray diffractions I_{111}/I_{220} and I_{200}/I_{220}, by which the degree of the atomic order for various x can be compared. In $0.1 \leq x \leq 1.8$ with increasing x the lattice constant a monotonically decreases with slightly concave curvature from a=0.588 to 0.568 nm. The monotonic change in a suggests that in these crystals Ru and Fe atoms are homogeneously distributed. As seen in Fig. 2(c), for $x \geq 1.0$ the degrees of both Y-Z and X-YZ orders decrease with increasing x, and eventually at x=1.8 the (111) peak disappears, which shows the vanishing of Y-Z order. This means that the crystal structure is $L2_1$ for $0.1 \leq x < 1.8$ and becomes $B2$ for x=1.8. This result suggests that Fe_2CrSi with the ordered structure may be unstable and is consistent with previous results that synthesizing stoichiometric Fe_2CrSi of single phase and of bulk form was unsuccessful.[18] With decreasing x from $x \sim 1.0$ these relative intensities of the superlattice peaks do not decrease and I_{200}/I_{220} even increases. Judging from our x-ray diffraction measurements, in the samples with nominally $0 \leq x \leq 0.05$, $L2_1$ structure is preserved in most part of the samples but minor impurity phases appear. Combining the fact that for $x \sim 0.1$ the lattice constant a does not depend much on x, solubility limit in this solid solution may exist. It is emphasized that from the x-ray data the degree of the atomic order does not decrease for $0.1 \lesssim x \lesssim 1.0$.

Figure 2(a) shows the results of low-temperature x-ray diffraction measurements for x=0.3. This sample exhibits a ferromagnetic transition at ~90 K and a subsequent transition with the decrease in M at ~40 K (see Fig. 3).[17] Within the experimental resolution no distinct change in the diffraction pattern was found at all temperatures and this shows that in this compound no structural transition occurs. High-temperature x-ray diffraction measurements up to 1073 K were performed for x=1.7.[19] The structure of this compound almost becomes $B2$ due to the decrease in Y-Z order. It exhibits the ferromagnetic transition at ~500 K. Within the experimental resolution, no change in the diffraction pattern including (111) peak was found up to 773 K. However, above 873 K there appear small but distinct extra peaks indicating impurities. This shows that above ~800 K minor fraction of the $L2_1$ phase begins to collapse. However, the indication of structural transitions was not found. Although measurements of the T dependence of x-ray patterns were carried out only for representative samples, it can be concluded that neither structural transition nor substantial change in the atomic order is induced by varying T and that the magnetic transitions in these compounds are not associated with structural transitions.

B. Magnetic properties

As reported previously,[17] the Fe-rich compounds has a ferromagnetic phase and, on the other hand, the Ru-rich compound (x=0.1) shows antiferromagnetic and SG behavior. Here we focus on the intermediate x region. Figure 3 shows the T dependences of magnetization $M(T)$ under μ_0H=1 T for $0.1 \leq x \leq 0.5$. For x=0.5 ferromagnetic behavior was observed, whereas $M(T)$ below ~50 K was found almost constant. More pronouncedly for x=0.3 a round maximum of $M(T)$ at ~40 K was observed. This round maximum turns to a clear peak at lower H, as shown in the inset of Fig. 3 for 0.1 T. We consider it a second magnetic transition and refer to the transition temperature as T_s. These results indicate that with decreasing T these compounds first become ferromagnetic and below T_s they turn to a state with some antiferromagnetic characters.

Figure 4(a) shows the magnetization versus magnetic field $M(H)$ ($-0.3 \leq \mu_0H \leq 0.3$ T) for x=0.5 at 4, 50, and 150 K.

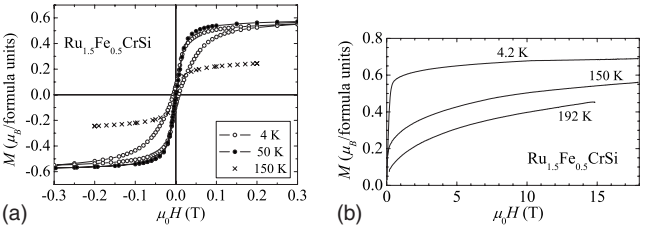

FIG. 4. Magnetic field dependence of magnetization for x=0.5. (a) In the low H range, hysteresis is observed at 4 K but not observed at 50 and 150 K. (b) High field magnetization up to 18 T.

In the $M(H)$ curves at 50 and 150 K no hysteresis was observed within the experimental resolution. The $M(H)$ at 4 K, however, shows clear hysteresis. Basically the same behavior of $M(H)$ was observed for $x=0.3$. These data demonstrate that there is a magnetic transition at T_s and that the magnetic state below T_s is ferromagnetic but different from the one at $T>T_s$. The shape of the hysteresis loop seems distorted as compared with typical $M(H)$ curves, that is, in spite of the considerable hysteresis the coercive field is small. The reason for this is not clear. However, considering the facts that the peak at T_s in $M(T)$ is round and an anomaly in $\rho(T)$ was not observed,[17] the transition at T_s seems to be a glassy one.

Figure 4(b) shows $M(H)$ for $x=0.5$ up to $\mu_0 H=18$ T at representative T. These data can be interpreted as those of ferromagnets. At 4.2 K M almost saturates to $0.7\mu_B$ above ~3 T. Following an initial rapid increase, neither nonlinear increase nor discontinuous jump in $M(H)$ was observed. Although M_0 for $x=0.5$ is smaller than that for $x\sim 2$, which is close to $\sim 2\mu_B$, H as high as 18 T does not recover M to this value.

As seen in Fig. 3, although there is considerable difference between the ferromagnetic behavior for $x=0.5$ and the antiferromagnetic and SG-type behavior for $x=0.1$, it appears that the $M(T)$ curves vary systematically and gradually with x in magnitude and in appearance. This continuity is also seen in the data at 0.1 T for $x=0.3$ shown in the inset of Fig. 3. These data resemble $M(T)$ at 1 T for $x=0.1$ (Ref. 17) and 0.2, which show a maximum and a hysteresis between ZFC and FC in the further lower-temperature range. For $x=0.3$ a drop in $M(T)$ below $T\sim 10$ K in ZFC even at 1 T suggests existence of hysteresis between ZFC and FC. On the other hand, the data for $x=0.1$ indicate the absence of ferromagnetism in spite of the continuity mentioned above and, moreover, the peak becomes sharper and the hysteresis is more pronounced. We confirmed from Arrott plots that there is no spontaneous magnetization for $x\leq 0.2$, while for $x\geq 0.3$ the feature of $M(H)$ and the Arrott plot show that spontaneous magnetization arises. Judging from these, for $x=0.1$ and 0.2, ferromagnetism indeed disappears. We tentatively regard the peak at $T=30$ K as an indication of an antiferromagnetic transition and refer to this temperature as T_N^*. Although it is clear that ferromagnetism disappears at low x, with decreasing x the ferromagnetic state seems to vanish gradually as mentioned above.

C. Spin-glass behavior

Figure 5(a) shows the temperature dependence of M in $\mu_0 H=1$ T for $x=0.1$. As described in Sec. III B, the peak in $M(T)$ at $T_N^*=30$ K was found and this can be seen as an indication of an antiferromagnetic transition. In the lower T-range hysteresis in $M(T)$ between ZFC and FC was found, and this indicates that in these compounds a SG state is realized. Moreover, a rapid decrease in M below ~5 K is seen only for ZFC. To clarify the thermodynamic nature specific heat C_p was measured. Figure 5(b) shows the result of C_p in zero magnetic field. With increasing temperature C_p increases monotonically. In spite of the clear peak in $M(T)$ at T_N^*, in $C_p(T)$ neither a peak nor an anomaly which indicates a

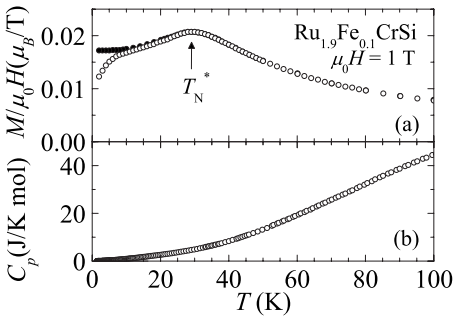

FIG. 5. Temperature dependence of the magnetization M and specific heat C_p of $Ru_{1.9}Fe_{0.1}CrSi$ are compared. (a) $M(T)$ in $\mu_0 H=1$ T under zero-field-cooling (open circles) and field-cooling (solid circles) conditions. (b) $C_p(T)$ in zero magnetic field. Although the peak in $M(T)$ is found at T_N^*, in $C_p(T)$ no anomaly is found at T_N^* or at any other temperatures.

phase transition was observed at around T_N^* or at any other temperatures. The result of $C_p(T)$ shows that there is no phase transition to a long-range-ordered state in $Ru_{1.9}Fe_{0.1}CrSi$.

In order to investigate the SG state of $Ru_{1.9}Fe_{0.1}CrSi$ we performed detailed measurements of M. Figure 6 shows $M(T)/\mu_0 H$ for ZFC and FC processes at various H, where the vertical scale is adjusted to the data of 1 T and the other zeros are each shifted for clarity. As shown in this figure, the peak in $M(T)$ appears at T_N^* and SG behavior, namely, the separation between the ZFC and FC curves, appears at a temperature lower than T_N^* as mentioned above. With increasing H from ~0.5 T the peak at T_N^* (=30 K at $\mu_0 H=1$ T) shifts slightly to lower T and becomes broader whereas still discernible at 5 T. A trace of the peak in $M(T)$ would remain beyond ~20 T as recognized from high-field $M(H)$ data in Fig. 7(a). It appears that at lower H below ~0.05 T there are two humps: a hump at $T\sim 18$ K and a

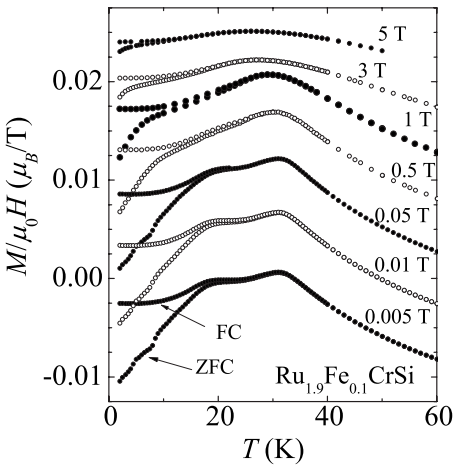

FIG. 6. Temperature dependence of magnetization divided by magnetic field $(M/\mu_0 H)$ of $Ru_{1.9}Fe_{0.1}CrSi$ at various magnetic fields in the low-temperature region. The vertical scale is adjusted to the data for 1 T. For the other fields, each vertical scale is shifted by 0.005 μ_B/T. Results for ZFC and FC conditions are shown. The separation of the magnetization between ZFC and FC in the low-temperature region is seen at all H.

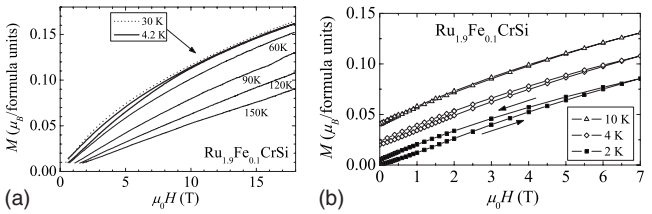

FIG. 7. (a) Magnetic field dependence of magnetization M of $Ru_{1.9}Fe_{0.1}CrSi$ in high field. Data between 4.2 and 40 K almost fall into a single curve identical to those for 4.2 and 30 K. The results for increasing and decreasing H at 4.2 K (thick line) are shown and hysteresis is observed. (b) $M(H)$ below 7 T at $T=2$, 4, and 10 K. The origins of M for 4 and 10 K are shifted by 0.02 and 0.04 (μ_B/f.u.), respectively. Considerable hysteresis is seen at $T \leq 4$ K in contrast to the only small hysteresis at 10 K.

hump at $T_N^* \sim 30$ K which we have called the peak. Although at higher H the hump at lower T seems to disappear, an abrupt downturn of M still remains only in the ZFC process, as we can see in a rapid decrease in $M(T)$ below ~ 5 K for 1 T. This downturn appears rather sharp. However, it is seen only in ZFC at higher H. This downturn is interpreted as the occurrence of strong irreversibility and seems to indicate a crossover to a different state.

This kind of successive changes in irreversibility behavior are seen in some ferromagnetic glassy systems where ferromagnetism and antiferromagnetism compete. Actually, for example, similar data were observed in low H ($\mu_0 H = 0.01$ T) in the Heusler compound Fe_2MnSi.[20] It is known that Fe_2MnSi exhibits a ferromagnetic transition and at a further lower temperature a second transition to a reordered phase with antiferromagnetic components.[21–23] The successive changes in irreversibility behavior were observed at low H in the reordered phase, where domain effects should play an important role. On the contrary, in the present compound ferromagnetism no longer exists and the hysteresis in $M(T)$ is seen even at relatively high H. Therefore the behavior observed for $x=0.1$ is considered not to originate from domain effects but as an intrinsically irreversibility phenomenon, and in this sense this is unique.

Figures 7(a) and 7(b) show the results of $M(H)$ for $x=0.1$ up to $\mu_0 H = 18$ T at various T and those up to 7 T at low T with increasing and decreasing H, respectively. At higher T above 100 K $M(H)$ curves are almost straight lines. Below 100 K it becomes a slightly concave curve, which corresponds to the deviation from the Curie-Weiss law in susceptibility.[17] Below ~ 30 K, where the peak in $M(T)$ is seen, the magnitude of M barely varies and in the scale of Fig. 7(a) the $M(H)$ curves at $4 \leq T \leq 40$ K almost merge into a single curve. As already pointed out, this indicates that the transition at $T_N^* \sim 30$ K sustains beyond $\mu_0 H = 18$ T. At all temperatures where the measurements were performed M is far from saturation even up to 18 T. Anomalies which show a spin-flop or a metamagnetic transition were not observed. These results strongly suggest antiferromagnetic nature at low T, although definite signs of the transitions in $M(H)$ were not observed.

As shown in Fig. 7(a) clear hysteresis was observed in $M(H)$ at 4.2 K. To examine in more detail the measurements

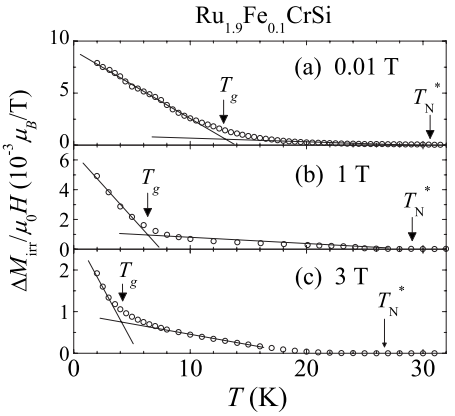

FIG. 8. Temperature dependence of $\Delta M_{\rm irr}/\mu_0 H$ for $x=0.1$ at (a) $\mu_0 H = 0.01$ T, (b) 1 T, and (c) 3 T. $\Delta M_{\rm irr}$ is irreversible magnetization defined by $\Delta M_{\rm irr}(T) = M_{\rm FC}(T) - M_{\rm ZFC}(T)$. T_N^* is peak temperature in $M(T)$. T_g is determined as the intersection of linear extrapolations as shown in these figures.

of $M(H)$ at low T were performed up to $\mu_0 H = 7$ T and the results are presented in Fig. 7(b). At lower T (=2 and 4 K) clear and relatively large hysteresis between increasing and decreasing H is seen. This hysteresis almost disappears at 10 K and the magnitude of M is almost unchanged by changing T in contrast to the change in the degree of hysteresis. This result is interpreted as the occurrence of the strong irreversibility of M below ~ 5 K, which is consistent with $M(T)$ measurements.

One may consider that the two humps in $M(T)$ seen in Fig. 6 originate from the inhomogeneity of the samples. For the following reasons we believe that our samples are homogeneous and the observed properties are intrinsic. First, from x-ray diffraction measurements the samples are single phase and homogenous in overall x range where we performed measurements. Second, the peak at $T_N^* = 30$ K and the downturn in ZFC at lower T are clear and systematically change with H. In particular, the peak and the downturn become sharper as H increases to ~ 1 T from lower H. Third, the $M(H)$ curves below $T_N^* = 30$ K is insensitive to T. This indicates that the most of the magnetic part in the samples turns into an antiferromagnetism-like state below T_N^*. Nevertheless, as shown in Fig. 7(b), in the $M(H)$ curves below 4 K considerable hysteresis appears. Therefore these two anomalies are hardly considered to originate from different parts of the sample.

It should also be pointed out that the magnitude of M was found independent of time within the period of our measurements, although in SG relaxation phenomena are often observed. Typically we took half a day to measure at a particular H and within this period any effects of time were hardly seen.

Another notice is that in the case for the presence of strong irreversibility like this, the shape of $M(T)$ curves would appear so different depending on T at which H is applied initially from $H=0$ in ZFC measurements. When we discuss the irreversibility of $M(T)$ curves, ZFC measurements were performed after cooling a sample under $H=0$ down to $T=2$ K and then applying H at this temperature.

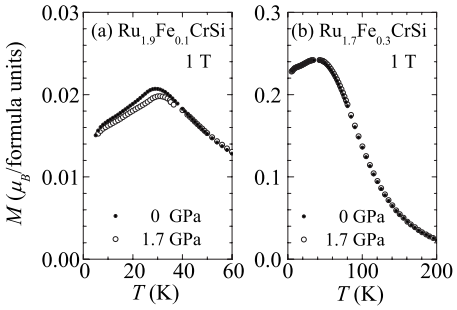

FIG. 9. Magnetization with pressures of 1.7 and 0 GPa under $\mu_0 H=1$ T for $x=0.1$ and 0.3.

To estimate the strength of the irreversibility in the SG state, $\Delta M_{\rm irr}(T)=M_{\rm FC}(T)-M_{\rm ZFC}(T)$ is defined as the irreversible magnetization. In Fig. 8 representative $\Delta M_{\rm irr}(T)/\mu_0 H$ are shown. We can divide into three temperature regions from this graph: the high-T region where $\Delta M_{\rm irr}$ is zero, the weak irreversibility region in the intermediate T range where $\Delta M_{\rm irr}$ is small, and the strong irreversibility region in the low-T range where $\Delta M_{\rm irr}$ exhibits rather a sharp rise.

As shown in Fig. 8 the onset T of the weak irreversibility appears to coincide just with T_N^* at lower H. At higher H the onset of irreversibility appears to be lower than T_N^*. However, $\Delta M_{\rm irr}/M$ is small at higher H and $\Delta M_{\rm irr}$ appears to approach zero asymptotically. Taking these into account, the determination of the onset of weak irreversibility is subtle and in this paper we suppose that the onset T of the weak irreversibility coincides with T_N^* and will not discuss this further.

The onset T of the strong irreversibility, which will be denoted by T_g, is determined as the intersection of the linear extrapolations of $\Delta M_{\rm irr}(T)/\mu_0 H$ from the strong and the weak irreversibility regions as shown in Fig. 8. Although this determination seems rather arbitrary, if we determine T_g as the maximum of the derivative of $M(T)$ in ZFC, for example, it makes little difference. This also demonstrates that the downturn in ZFC in higher H and the hump at lower T in lower H in $M(T)$, shown in Fig. 6, are connected and the downturn can be interpreted as the occurrence of strong irreversibility. Anyway, this boundary should be considered as a crossover. The $M(H)$ phase diagram is constructed by plotting T_N^* and T_g and shown in Fig. 11.

D. Pressure effect

We performed the measurements of the pressure effect on magnetic properties for $x=0.1$, 0.3, and 0.5. Figure 9 shows $M(T)$ at 1 T with pressures of 0 and 1.7 GPa for $x=0.1$ and 0.3. It seems that for $x=0.3$ M is not affected by pressure up to 1.7 GPa. For $x=0.5$ the same result that $M(T)$ does not depend on pressure was obtained. On the other hand, for $x=0.1$ the peak at T_N^* shifts to higher temperature by 2 K and around the peak the magnitude of M shifts to smaller value with applying a pressure of 1.7 GPa. From these results it is concluded that the pressure of 1.7 GPa hardly influences the ferromagnetic properties while it has some effects of enhancing the antiferromagnetic properties. If we assume that pressure effect is equivalent to volume contraction, in this series of the compounds pressure corresponds to the increase in x [see Fig. 2(b)] and this would enhance ferromagnetism. This is not consistent with the present results and thus the results cannot be simply interpreted. Nevertheless, in Heusler and half-Heusler compounds the increase in magnetic transition temperatures with pressure was reported and $\Delta T_N^*/\Delta P=1.2$ K/GPa in the present measurements is comparable to these data.[24,25]

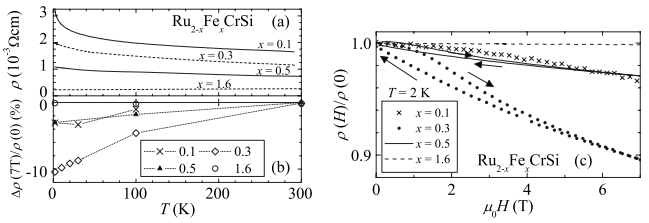

FIG. 10. (a) Temperature dependence of electrical resistivity ρ for representative x. (b) Magnetoresistance in $\mu_0 H=7$ T, $\Delta\rho(7\ \mathrm{T})/\rho(0\ \mathrm{T})$, at several temperatures is shown for representative x $[\Delta\rho(7\ \mathrm{T})=\rho(7\ \mathrm{T})-\rho(0\ \mathrm{T})]$. (c) H dependence of ρ at 2 K in the form of $\rho(H)/\rho(0\ \mathrm{T})$ for representative x. For $x=0.3$ and 0.5 hysteresis is observed and the arrows show the direction of varying H.

E. Magnetoresistance

In Fig. 10(a) the T dependences of electrical resistivity $\rho(T)$ at $H=0$ for representative samples are shown. As reported in the previous paper a semiconductor-metal crossover in $Ru_{2-x}Fe_xCrSi$ was found around $x\sim 0.7$.[17] To study the transport properties further magnetoresistance (MR) measurements were performed up to 7 T. Figure 10(c) shows H dependence of ρ at 2 K in the form of $\rho(H)/\rho(H=0)$ for representative x. For Fe-rich metallic samples ($x=1.5$ and 1.6) $\rho(H)$ is independent of H. For intermediate x samples ($x=0.3$ and 0.5) fairy large negative MR was observed. Moreover, hysteresis in $\rho(H)$ at 2 K was observed and seems in accordance with that of $M(H)$, and this type of behavior is reminiscent of granular type MR effect.[26] With increasing magnetic field $\rho(H)$ shows tendency to saturation. These results indicate that MR is related to M. The Ru-rich ($x=0.1$) compound exhibits also fairy large negative MR, whereas the dependence on H is different. In this case $\rho(H)$ does not exhibit tendency to saturation up to 7 T and appears to exhibit almost H^2 dependence. Moreover hysteresis in $\rho(H)$ was not observed. These results suggest that the origin of MR for $x=0.1$ is different from that for $x=0.3$ and 0.5.

Figure 10(b) shows $\Delta\rho(7\ \mathrm{T})/\rho(0\ \mathrm{T})$ $[\Delta\rho(7\ \mathrm{T})=\rho(7\ \mathrm{T})-\rho(0\ \mathrm{T})]$ at various T obtained from $\rho(H)$ measurements. For Fe-rich metallic samples MR is practically zero for all T and H ranges where measurements were made. When MR is observed in the present series of compounds, it is always negative. The magnitude of MR increases with decreasing T. The maximum of the negative MR ratio $\Delta\rho(7\ \mathrm{T})/\rho(0\ \mathrm{T})$ is 10% for $x=0.3$ at 2 K.

IV. DISCUSSION

A. Spin-glass state

Next we consider the nature of the transitions at T_N^* and T_g and the SG state. The dependences of M on T and H indicate

that the transition at T_N^* has a character of usual antiferromagnetic transitions. This is consistent with the theory which shows that for $x \leq 0.25$ an antiferromagnetic state is energetically preferable to the ferromagnetic state.[14] It was reported that an isoelectronic structure compound Ru_2CrGe is an antiferromagnet with the Néel temperature of 13 K.[27] Furthermore, the reported ferromagnetic properties of $Ru_{2-x}Fe_xCrGe$ $(0.25 \leq x \leq 1.5)$ (Ref. 28) are very similar to those of $Ru_{2-x}Fe_xCrSi$. In addition, in $Ru_{1.9}Fe_{0.1}CrGe$ similar SG behavior to that of $Ru_{1.9}Fe_{0.1}CrSi$ has been found.[19] Thus it can be considered that $Ru_{2-x}Fe_xCrGe$ is an analogous series to $Ru_{2-x}Fe_xCrSi$. And also in Mn-based Heusler compounds including Ru, antiferromagnetic orders were reported.[29,30] Unfortunately we could not obtain single-phase sample of Ru_2CrSi; it would be an antiferromagnet, if it exists. Considering these facts, the transition at T_N^* might be regarded as an antiferromagnetic transition. And in this case the transition at T_g should be regarded as a re-entrant-type SG transition. Re-entrant-type SG transitions were reported for both ferromagnetic and antiferromagnetic cases in the Ising-type materials $Fe_xMn_{1-x}TiO_3$.[31]

However, by $\rho(T)$ measurements an anomaly at T_N^* and also at T_g could not be detected.[17] In addition, as shown in Fig. 5(b), the specific heat does not exhibit any peak or anomaly at T_N^*, or at any other temperatures. These results clearly demonstrate that there is no phase transition to a true long-range-ordered antiferromagnetic phase in $Ru_{1.9}Fe_{0.1}CrSi$. We thereby guess another plausible possibility that there are SG transition at T_N^* and another successive SG transition at a lower temperature T_g.

Successive SG transitions were investigated both theoretically and experimentally. It was shown theoretically that in an Ising SG model there is a single SG transition under H and the boundary is called the de Almeida-Thouless (AT) line.[32] This theory was generalized to a Heisenberg model and it was shown that there are successive SG transitions in finite H.[33,34] With lowering T from a paramagnetic phase, first the freezing of the transverse components of M occurs, and this boundary is called the Gabay-Toulouse (GT) line. Further lowering T, then, the freezing of the longitudinal components of M occurs and this boundary is called the AT line in the Heisenberg case. In this model the two boundaries meet at $H=0$. This phase diagram was experimentally investigated.[35,36] If anisotropy is taken into account, it was shown theoretically that there can be successive SG transitions even at $H=0$ for a certain range of anisotropy[37] and experiments using single crystals of SG materials supported this theory.[38] Besides, in the presence of a ferromagnetic order, the existence of successive SG-like transitions was proposed theoretically.[33,34]

In the light of the above knowledge, the SG behavior of $Ru_{1.9}Fe_{0.1}CrSi$ seems to be explained by a following model qualitatively: a certain weak anisotropy or ferromagnetic interaction (possibly a short-range order) exists and causes AT- and GT-like successive SG transitions.

We compare the phase boundary of T_g shown in Fig. 11 with the AT line. The phase boundary of $T_{AT}(H)$ for small field is expressed as

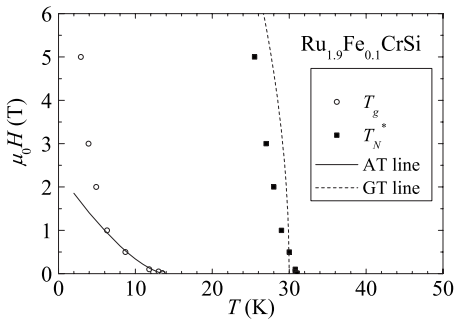

FIG. 11. H-T phase diagram of $T_g(H)$ and $T_N^*(H)$ for $Ru_{1.9}Fe_{0.1}CrSi$. The solid line shows $[T_g(0)-T_g(H)]^{3/2}$ dependence which is characteristic of AT boundary. The dashed line shows $[T_N^*(0)-T_N^*(H)]^{1/2}$ dependence which is characteristic of GT boundary.

$$\mu_0 H = A_{AT}\{1 - T_{AT}(H)/T_{AT}(0)\}^{3/2}. \quad (1)$$

For three-dimensional Heisenberg system with spin S and A_{AT} is given by

$$A_{AT} = \sqrt{\frac{2}{5}}\frac{k_B T_{AT}(0)}{g\mu_B S}. \quad (2)$$

In Fig. 11 the solid line represents Eq. (1) with $A_{AT}=2.3$ T and $T_{AT}(0)=13.8$ K, which are determined so that the line well reproduces the experimental results of T_g for $\mu_0 H \lesssim 1$ T. While coefficient A_{AT} is calculated to be 8.1 T from Eq. (2) with $T_{AT}(0)=13.8$ K and $gS=1.6$, which is chosen because this value of the local moment is expected from the Curie-Weiss law [see Fig. 12(b)]. Experimentally obtained A_{AT} is smaller than but comparable to the theoretical one. In previous studies experimentally obtained A_{AT} is often much smaller than the theoretical value while good agreement with the theoretical value was also reported.[36] As seen in Fig. 11 the data deviate from the line of Eq. (1) in high H range. This is not strange because Eq. (1) is an approximate expression for low H. Above all in the experiments this transition is characterized by the onset of strong irreversibility of M. It was pointed out in the theory that strong irreversibility is a characteristic of the AT transition.[34] And in the experiments at higher H for ZFC the anomaly is remarkable at T_g while for FC the anomaly is not noticeable around T_g. This behavior looks like the so-called pin effect. Therefore it should be considered as a crossover rather than an ordinary phase transition. It was pointed out previously that an AT transition is a crossover.[34,35] Hence the boundary $T_g(H)$ can be interpreted as a AT boundary.

As shown in Fig. 11 the change in T_N^* with H is small and might be small even up to higher H range beyond ~20 T. This kind of robustness to H is a character of GT line. We thus attempt to compare T_N^* with a theoretical GT line. The phase boundary of $T_{GT}(H)$ is expressed theoretically as

$$\mu_0 H = A_{GT}\{1 - T_{GT}(H)/T_{GT}(0)\}^{1/2}. \quad (3)$$

For three-dimensional Heisenberg system A_{GT} is given by

$$A_{GT} = \frac{4.3 k_B T_{GT}(0)}{g\mu_B S}. \quad (4)$$

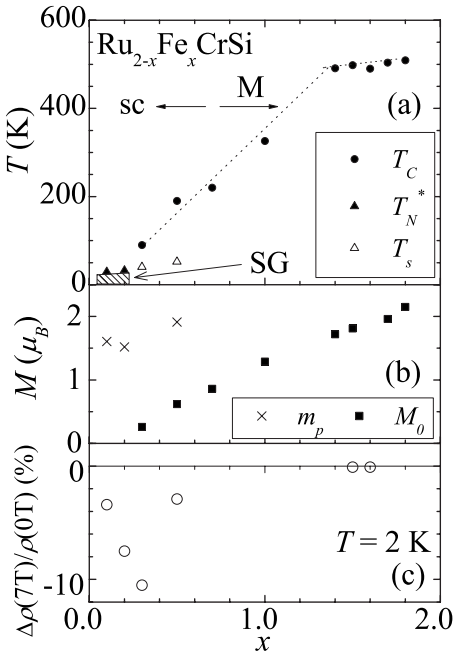

FIG. 12. (a) T-x magnetic phase diagram of $Ru_{2-x}Fe_xCrSi$. Curie temperature T_C, second magnetic transition T_s (x=0.3 and 0.5), and higher (antiferromagneticlike) SG transition T_N^* are shown. A metal (m)-semiconductor (sc) crossover is around $x \sim 0.7$. (b) x dependences of the saturation magnetization at 0 K M_0 and of the paramagnetic moment m_p deduced from the Curie-Weiss law. (c) Magnetoresistance in $\mu_0 H$=7 T, $\Delta\rho(7\ \mathrm{T})/\rho(0\ \mathrm{T})$, at T=2 K as a function of x.

Using experimentally determined A_{AT}=2.3 T with A_{GT}/A_{AT}=7.2 [estimated from Eqs. (2) and (4), with $T_{AT}(0)$=13.8 K and $T_{GT}(0)$=30 K], A_{GT} becomes 16.8 T. The dotted line drawn in Fig. 11 shows the GT line calculated by Eq. (3) using this A_{GT} with $T_{GT}(0)$=30 K. Although the plotted T_N^* at first glance does not appear to show $[T_N^*(0)-T_N^*(H)]^{1/2}$ dependence, considering that the broad transition prevents from determining $T_N^*(H)$ precisely, the data seem to reflect a characteristic of GT transition. Therefore the scenario that these transition lines are interpreted as AT and GT transitions is likely to be reasonable. However, at present we do not have precise microscopic information on the magnetic interactions and the anisotropy of this compound, and probably the magnetic interactions are more complicated than those postulated in theories. Thereby it is not clear that these theories directly apply to this material; nevertheless, it gives a qualitative view.

B. Magnetic phase diagram

Figure 12(a) shows the T-x magnetic phase diagram. Figure 12(b) shows the x dependence of M_0, which is determined from extrapolation of $M(T)$ at $\mu_0 H$=1 T to T=0 K, and paramagnetic moment m_p, which is evaluated from the effective magnetic moment m_{eff} determined from the Curie constant $[m_{\mathrm{eff}}=\sqrt{m_p(m_p+2)}]$, so that it can be compared directly with spin values (see, for example, Ref. 39). For $0.1 \leq x \leq 0.5$ m_p is $1.5\mu_B$–$1.9\mu_B$; this value is smaller than the value of $3\mu_B$ expected for Cr^{3+}. These values have ambiguity because the simple Curie-Weiss law does not necessarily hold for adequately wide temperature range.[17]

The value of M_0 for $x \simeq 2$ is nearly $2\mu_B$, which is theoretically expected. Therefore these compounds have been revealed to be candidates for materials with high spin polarization. It is also pointed out that the change in $M(T)$ curve seems sharp around T_C as in usual uniform ferromagnets. Judging from x-ray diffraction measurements the atomic order decreases with increasing x and the structure turns to $B2$ type at x=1.8. However, the magnetic properties change smoothly with x and do not seem to be influenced by atomic disorder.

As x decreases from $x \simeq 1.4$ to 0.3, M_0 and T_C decrease monotonically. This behavior of M_0 versus x differs from the theory which has shown that M_0 is nearly $2\mu_B$ and independent of x for all the ferromagnetic compounds. The reasons for the discrepancy can be considered as following.

First, atomic disorder affects the magnetism, in particular, half metallicity.[7–9,13,14] However, as mentioned above, Y-Z disorder in fact has little effect on the magnetism and the atomic order does not decrease for lower x. For this reason, it is concluded that atomic disorder is not a main reason for the discrepancy.

Second, itinerancy should be considered. The $4d$ state of Ru is extended; thereby, when Ru is increased, it is natural to think that itinerancy increases. With decreasing x the ratio M_0/m_p becomes considerably smaller. The decrease in M_0 might be considered as the effect of the increase in itinerancy.[16,39] However, it was reported that the analogous compound Ru_2CrGe has the magnetic moment of $1.45\mu_B$ at Cr in the antiferromagnetic phase.[40] This fact suggests that also $Ru_{2-x}Fe_xCrSi$ probably has the local moment of comparable size at Cr even for small x, and thus the decrease in M_0 does not necessarily mean the decrease in the local moment. Additionally the experiments revealed that increasing Ru in this series changes the compound from a metal to a semiconductor. This is not consistent with the increase in itinerancy. The theory demonstrates that M_0 is almost $2\mu_B$ for a wide range of x and that most part of the magnetic moment is attributed to Cr atoms, as it is known in Heusler compounds X_2YZ that most of the moment is usually carried by Y atom. For small x the value of m_p experimentally obtained is comparatively close to $2\mu_B$. Taking these into account, it is appropriate that we think it as a starting point that the Cr atoms carry the local magnetic moment of nearly $2\mu_B$ for all x in this series, as implicitly already assumed. However, the effect of itinerancy is obviously present and the itinerancy is likely to increase as Ru increases. Nevertheless the metal-semiconductor crossover occurs with increasing Ru. Although the electronic correlations might play a role, at present the origin is puzzling and remains a future problem.

Finally, the competition between ferromagnetism and antiferromagnetism is discussed and we consider it as a main reason. The appearance of SG for small x indicates the existence of several competing ferromagnetic and antiferromagnetic interactions. As shown in Fig. 12(a), for intermediate x (=0.3 and 0.5) the ferromagnetic transition at T_C and a second transition at a lower temperature T_s exist. For $T<T_s$ glassy ferromagnetism is realized as seen in the hysteresis in

M shown in the inset of Fig. 3 and in Fig. 4(a). Even for $T > T_s$ the ferromagnetic state may be different from that for higher x. This can be seen in the very gradual transition in the $M(T)$ curve around T_C under $\mu_0 H = 1$ T shown in Fig. 3.

One of the possible models to interpret these results for intermediate x is that the competition between ferromagnetic and antiferromagnetic interactions results in a magnetically inhomogeneous state, i.e., formation of ferromagnetic clusters in other nonmagnetic areas, which may turn into the spin-glass state for $0.3 \leq x \leq 0.5$. This model qualitatively explains not only the decrease in M_0 with decreasing x but the whole T-x phase diagram and other magnetic and magnetotransport properties. It is noted that the atomic order does not at least decrease in this range of x where the glassy magnetism appears.

For intermediate x the ferromagnetic moment is attributed to the ferromagnetic clusters which occupy partial fraction of the sample, and with decreasing x the reduction in the ferromagnetic clusters causes the decrease in M_0. In the other areas large magnetic moment is not induced even in high H range [Fig. 4(b)]. The formation of the clusters is also consistent with the gradual ferromagnetic transition observed in $M(T)$ under 1 T for intermediate x, which is in contrast to the transitions at T_C for higher x. In addition, it appears that with decreasing x the ferromagnetism fades out gradually around $x \sim 0.2$. This suggests that the disappearance of the ferromagnetism with decreasing x corresponds to breaking the connection of the ferromagnetic clusters. As shown in Fig. 12(a), the fact that T_s appears to connect continuously to T_N^* also suggests that for $x \geq 0.3$ SG regions already coexist outside the ferromagnetic clusters.

Then we turn to MR of this system. For intermediate x at low temperatures the ferromagnetic clusters form domain structure and this may increase the role of spin-dependent grain-boundary scattering, and this would lead to the fairy large MR and hysteresis behavior with varying H, as seen in Fig. 10(c). This behavior is different from that for the Fe-rich compounds ($x \geq 1.5$) in which no MR is observed. This also suggests that the Fe-rich compounds are usual uniform ferromagnets. Figure 12(c) shows x dependence of MR ratio in $\mu_0 H = 7$ T, $\Delta\rho(7\ \mathrm{T})/\rho(0\ \mathrm{T})$, at 2 K. MR is the largest at $x = 0.3$ and fairy large at $x = 0.1$. As mentioned above, MR for $x = 0.1$ has different H dependence from that for $x = 0.3$ and no hysteresis. This means that for $x = 0.1$ the origin of negative MR, which may be in magnetic scattering, is different from that for intermediate x and again indicates that the ferromagnetic clusters disappear and the SG state occupies all over the compound. The similar magnetic and transport properties and T-x phase diagram (SG and adjacent nonuniform ferromagnetic phases) have been reported in other materials such as Co oxides.[26,41,42]

As already pointed out, Heusler compounds should inherently have tendency to magnetic frustration. However, thus far this viewpoint seems to have been overlooked. One of other examples in Heusler compounds where ferromagnetic and antiferromagnetic interactions exist is Fe_2MnSi, which exhibits the ferromagnetic transition and the second reordering transition at a lower temperature.[21–23] The substitution of V or Co suppresses the second transition and increases the ferromagnetic transition temperature.[43,44] This behavior seems similar to that of $Ru_{2-x}Fe_xCrSi$. However, in the case of Fe_2MnSi, both the ferromagnetic transition and the second reordering one were detected definitely by the measurements of M and ρ. The peak in specific heat at the reordering transition was also reported.[45] This means that these transitions of Fe_2MnSi are undoubtedly phase transitions. Thereby, although it is certain that there are competing interactions in both the materials, the natures of magnetism are distinctly different and this fact is one of the examples which demonstrate the diversity of magnetism in Heusler compounds.

V. CONCLUSIONS

We previously revealed that for larger x ($\gtrsim 1.5$) $Ru_{2-x}Fe_xCrSi$ is ferromagnetic with Curie temperature of ~ 500 K and its saturated magnetic moment is close to the theoretically predicted value. This means that the Fe-rich compound is a candidate for a material with high spin polarization and potentially useful for spintronics. The Ru-rich compound ($x = 0.1$) exhibits in the temperature dependence of magnetization a peak, which might indicate an antiferromagnetic transition, and irreversible behavior at lower temperatures, which suggests appearance of a spin-glass state. In the present work we have performed specific-heat and magnetization measurements and have revealed that for $x = 0.1$ there is no antiferromagnetic transition. With further decreasing temperature the occurrence of strong irreversibility in magnetization has been observed. We have proposed that these results can be interpreted as successive spin-glass transitions. For intermediate x (=0.3 and 0.5) glassy ferromagnetism has been observed. The magnetic and the magnetoresistive properties of these compounds can be explained by assuming the formation of ferromagnetic clusters in the other nonmagnetic region, which will turn into the spin-glass state at lower temperatures. These results show that a few ferromagnetic and antiferromagnetic interactions compete in this series for lower x. The Heusler compounds $Ru_{2-x}Fe_xCrSi$ have found to be a noticeably interesting system where a variety of phenomena are caused by the competition of magnetic interactions.

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