Abnormal magnetoresistance in ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ antiferromagnets

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The magnetic, transport and magnetotransport properties of antiferromagnetic ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ ($x=0.17$ and 0.2) compounds are investigated systematically. For both compounds, a large positive magnetoresistance (MR) is observed in the temperature regime below the temperature (T_t) for a transition from a triangular to a collinear antiferromagnetic spin configuration, while a negative magnetoresistance appears between T_t and the Néel temperature T_N, and then the MR ratio turns to be positive again above T_N. Below T_t, the positive MR reaches a maximum ratio of 6.1% at 5 K at an applied magnetic field of 5 T for the ε-$(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ compound. For the ε-$(Mn_{0.83}Fe_{0.2})_{3.25}Ge$ compound, the positive MR ratio reaches its maximum at 120 K and nearly vanishes at 5 K. The origin of the anomalous positive and negative MR effects is discussed in terms of the shrinkage of the orbits, the spin fluctuations, the spin configurations, and the magnetic transitions.

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I. INTRODUCTION

The intermetallic compound ε-Mn_3Ge crystallizes in the hexagonal Ni_3Sn type (DO_{19}) (space group $P6_3/mmc$), in which Mn positions are at $z=1/4$ and $3/4$.[1] The magnetization of ε-Mn_3Ge compound was measured by Ohoyama,[2] and a possibility of a feeble ferromagnetism parasitic on antiferromagnetism was mentioned. Kouvel and Kasper,[3] Kádár and Krén,[4] and Tomiyoshi[5] proposed a triangular spin configuration on the basis of neutron powder diffraction measurements, and the weak ferromagnetism observed up to 360 K was brought by deflection, within the basal plane, of individual magnetic moments toward their own easy axes from the regular triangular configuration. The isostructural compound ε-Fe_3Ge is ferromagnetic below its Curie temperature of 638 K.[6] Hori *et al.*[7] and Niida *et al.*[8] investigated the magnetic phase diagram for the pseudobinary system of ε-$(Mn_{1-x}Fe_x)_{3+\delta}Ge$ with $0 \leqslant x \leqslant 1$. Complicated magnetic structures, including ferromagnetic structure and two kinds of antiferromagnetic structures, appear in ε-$(Mn_{1-x}Fe_x)_{3+\delta}Ge$ compounds. At the magnetic transition between two antiferromagnetic configurations, the magnetization and electrical resistivity were abruptly changed.[8,9] Usually, sharp changes in physical properties such as electrical resistivity and magnetization of bulk materials caused by metamagnetic transitions can lead to a large magnetoresistance (MR). The previous studies focused on the magnetic properties, especially emphasizing the effect of the composition on magnetic transitions. Little work on the transport properties and no work on the magnetotransport properties have been done for the ε-$(Mn_{1-x}Fe_x)_{3+\delta}Ge$ compounds. In particular, we want to explore if abnormal magnetotransport properties can appear at the abrupt change of physical properties in this system. The emphasis of this work is on the relation between the magnetic transitions, anomalous transport behaviors, and magnetoresistance of the antiferromagnetic ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ ($x=0.17$ and 0.2) compounds.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of ε-$(Mn_{1-x}Fe_x)_{3+\delta}Ge$ ($\delta=0.25$ and $x=0.17$ and 0.2) compounds were prepared by arc melting Mn (99.9%), Fe (99.9%), and Ge (99.999%) under a high purity atmosphere. An excess of Mn (5 wt %) was added to compensate for weight loss during melting. The ingots were ground pulverized into powders and pressed into pellets. The pellets were homogenized in an evacuated and sealed silica tube at 800 °C for four days, and then quenched in water. X-ray diffraction studies were carried out at room temperature with Cu $K\alpha$ radiation in a $D/\max$-γA diffractrometer with a graphite crystal monochromator, which certified that the sample displayed peaks characteristic of hexagonal DO_{19} type. The magnetic properties were measured using a superconducting quantum interference device magnetometer in the temperature range from 5 to 360 K. The polycrystalline sample was cut into a bar shape ($1\times1\times8$ mm^3) for measurements of transport properties. Magnetotransport properties were measured using a standard four-probe method with a dc perpendicular to magnetic fields. Since the first-order nature of the magnetic transitions is not the focus of this work and the thermal hysteresis observed in our work is small,[10,11] we represent only the data recorded during heating.

III. RESULTS AND DISCUSSION

A. Magnetic and transport properties

Figure 1 represents the temperature dependence of magnetization and electrical resistivity of ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ compounds, with $x=0.17$ and 0.2, respectively. ε-$(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ undergoes two successive transitions at $T_t \sim 90$ K [antiferromagnetic II (AFII)—antiferromagnetic I(AFI)] and $T_N \sim 240$ K (antiferromagnetic I–paramagnetic) as shown in Fig. 1(a). Same transitions at $T_t \sim 130$ K and $T_N \sim 214$ K were observed for ε-$(Mn_{0.8}Fe_{0.2})_{3.25}Ge$, as seen in Fig. 1(b). The discontinued change of magnetization at T_t for both compounds can be explained by the change of the two different magnetic states. According to neutron diffraction, Hori *et al.*[7] assumed that there were two antiferromagnetic phases AFI and AFII, respectively, above and below T_t. AFI was arranged as a triangular antiferromagnetic spin con-

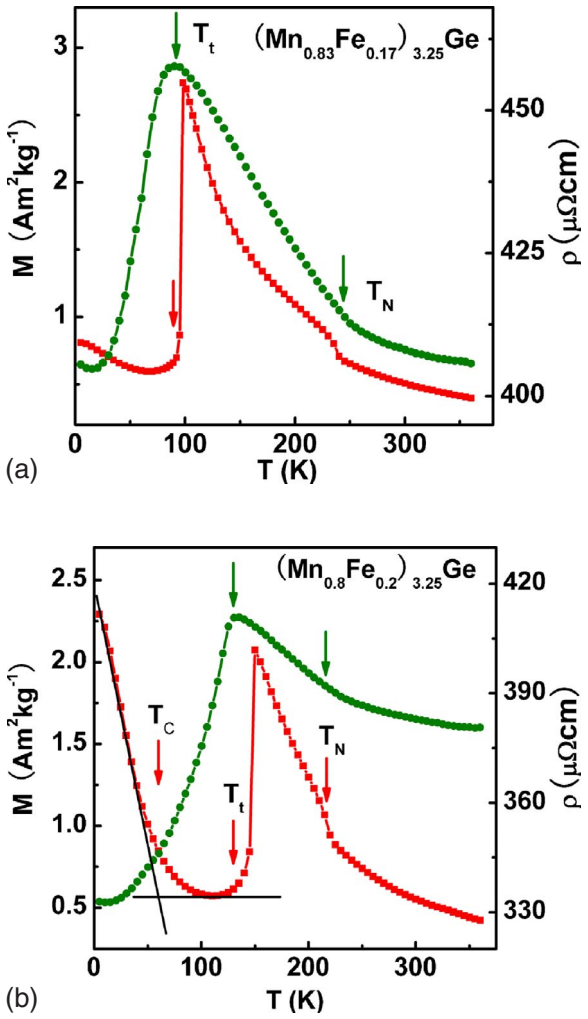

FIG. 1. (Color online) Temperature dependence of the resistivity (circles) and the magnetization (squares) of (a) $(Mn_{0.83}Ge_{0.17})_{3.25}Ge$ and (b) $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$. The resistivity was measured at zero field and the magnetization was measured at a magnetic field of 0.95 T.

figuration in the *a-b* plane, while AFII had a collinear-type antiferromagnetic configuration. In AFII, the moments of Mn and Fe atoms were ferromagnetically coupled in the *a-b* plane and antiferromagnetically coupled between ferromagnetic sheets. The weak ferromagnetism in the AFI phase was due to the deviation of individual magnetic moments toward their own easy axes from the regular triangular configuration. Therefore, the magnetization above T_t is higher than that below T_t. The dramatic increase of magnetization of $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ below T_t [as shown in Fig. 1(b)] is due to the appearance of ferromagnetism with increasing Fe concentration, which is consistent with the previous study.[7] The Curie temperature T_C is 60 K as determined by the crossing point of two tangent lines of the magnetization curve shown in Fig. 1(b).

For both $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ and $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ compounds, the overall behavior of the resistivity is semiconductor like. The slope of the resistivity changes obviously at and/or below T_N but changes abruptly at and/or below T_t, as shown in Figs. 1(a) and 1(b). The degree of the magnetic

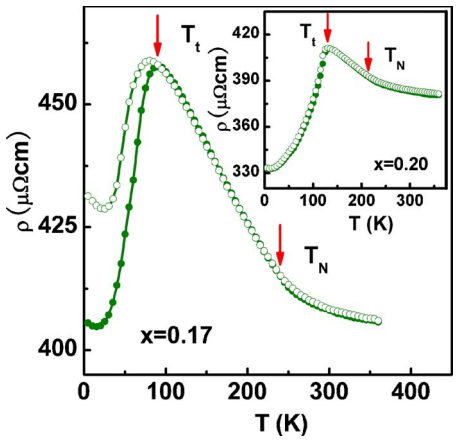

FIG. 2. (Color online) Temperature dependence of the resistivity of $(Mn_{0.83}Ge_{0.17})_{3.25}Ge$ at zero field (solid) and at a magnetic field of 5 T (open). The inset is that of $(Mn_{0.8}Ge_{0.2})_{3.25}Ge$.

ordering in the collinear antiferromagnetic phase AFII is higher than that in the triangular phase AFI, which can be confirmed by the positive magnetic entropy change ΔS_m obtained around T_t.[10] Thus, the electrons suffer less magnetic scattering probability in AFII phase than in AFI phase, resulting in the decrease of the electron resistivity at and/or below T_t. In a previous study,[8] the unit cell volume of the ε-$(Mn_{0.85}Fe_{0.15})_{3.1}Ge$ compound decreased by 2% at T_t. It is known that the resistivity is sensitive to the density of states (DOS) of electrons near the Fermi surface. The decrease of the unit cell may lead to an increase of the DOS near the Fermi surface and thus a decrease of the resistivity. Both the two factors mentioned above can influence the conductivity and lead to the abrupt decrease of resistivity below T_t. There exists a resistivity minimum at 15 and 10 K, respectively, for both compounds. The resistivity minimum is the indication of the end of the magnetic transition. The temperature range of the magnetic transition (120 K, from 130 to 10 K) for $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ is wider than that (75 K, from 90 to 15 K) for $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ because of the increase of the inhomogeneity, caused by increasing the Fe concentration.[9]

B. Magnetotransport properties

As seen from the results above, the magnetic transition has a prominent influence on the transport properties for $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ and $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ compounds. Then, it is interesting to study the magnetotransport properties of the compounds. The temperature dependence of the resistivity of ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ (x=0.17 and 0.2) compounds at zero field and at a magnetic field of 5 T is given in Fig. 2. Anomalous MR effect $[\Delta\rho/\rho=[\rho(H)-\rho(0)]/\rho(0)]$ is observed for $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$. Namely, a substantial positive MR appears below T_t, a small negative MR is observed between T_t and T_N, and then MR turns to be positive again above T_N. The anomalous MR effect can be clearly seen in the upper panel of Fig. 3(a). For $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$, the positive MR increases rapidly below T_t, reaches a value of 6.1% at 5 K (in a field of 5 T), and gives no sign of saturation up to 5 T. The negative MR between T_t and T_N, and the

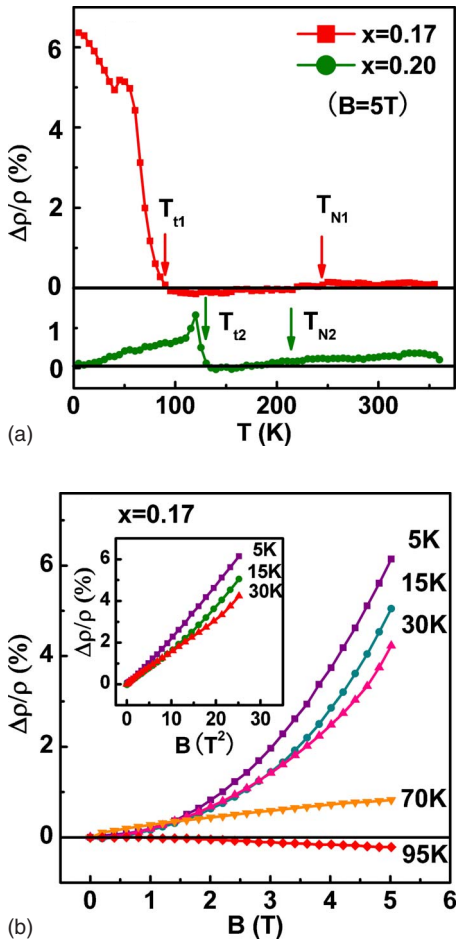

FIG. 3. (Color online) (a) Temperature dependence of MR $(\Delta\rho/\rho)$ at a magnetic field of 5 T for $(Mn_{0.83}Ge_{0.17})_{3.25}Ge$ and $(Mn_{0.8}Ge_{0.2})_{3.25}Ge$, (b) The magnetic field (H) dependence of MR $(\Delta\rho/\rho)$ at 5, 15, 30, 70, and 95 K [the inset shows the H^2 dependence of MR $(\Delta\rho/\rho)$ at 5, 15, and 30 K] for $(Mn_{0.83}Ge_{0.17})_{3.25}Ge$.

positive MR above T_N are very small. Same phenomenon is observed for the $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ compound, as seen in the inset of Fig. 2 and lower panel of Fig. 3(a). Different from $(Mn_{0.83}Fe_{0.17})Ge_{3.25}$, with decreasing temperature, the positive MR value increases below T_t, reaches a maximum at 120 K, and then nearly vanishes at 5 K. In order to confirm the anomalous MR effect, the variation of the resistivity with increasing magnetic field was measured at different temperatures for the most responsive $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ compound, and the results were represented in Fig. 3(b). At low temperatures (5, 15, and 30 K), the value of $\Delta\rho/\rho$ has strong quadratic dependence on the magnetic field H as shown in the inset of Fig. 3(b). As the temperature is close to T_t (70 and 95 K), the value of $\Delta\rho/\rho$ has a linear dependence on H (see Fig. 3).

If a material is nonmagnetic, its magnetoresistance should have the same sign for the whole testing temperature. However, for a magnetic alloy, anomalous MR can appear due to spin-mediated scattering. The electrical resistance is reduced when there is less scattering of the conduction electrons by the spins of the localized electrons. To the extent that an external field reduces the fluctuation of the local spins, the scattering will be reduced, which results in a negative MR. This mechanism of spin dependence of scattering can explain the negative MR appearing between T_t and T_N for both compounds. Negative MR can be observed also for triangular antiferromagnets $(Mn_{1-x}Fe_x)_{3.25}Ge$ with $x<0.17$.[11]

Usually, the positive MR is understood in terms of the Lorentz contribution to resistivity in the presence of a magnetic field H. The Lorentz contribution gives rise to a large positive MR only when the condition $\omega_c\tau \gg 1$ is satisfied (where ω_c and τ are the cyclotron frequency and conduction electron relaxation time, respectively). This condition holds well for extremely pure metallic single crystals at very low temperatures (where τ is very large, which, in turn, implies that $\rho \approx 10^{-2}\mu\Omega$ cm or less) and at large H (when ω_c is large). For our $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ and $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ compounds, the resistivity is large below T_t and above T_N, and τ is so small that even at large fields the above condition cannot be satisfied. Magnetoresistance can also be found to be positive due to intrastate correlation on variable range hopping (VRH), according to the theoretical calculation by Kurobe and Kamimura.[12] It shows a linear magnetic-field dependence in lower magnetic fields and saturates above a certain magnetic field. Its MR is usually observed at extremely low temperatures. This mechanism cannot explain the quadratic and linear H dependences of positive MR (which gives no sign of saturating) below T_t and above T_N.

The resistivity of $Mn_{3.25}Ge$ presents a typical metallic conductivity behavior. However, in our study,[11] the resistivity of the Fe-containing $(Mn_{1-x}Fe_x)_{3.25}Ge$ compounds exhibits anomalous semiconductorlike behavior. This metal-insulator transition is due to disorder, originated from Fe substitution for Mn. As the electrons are strongly localized, the conductivity can be described by VRH conductivity. The temperature dependence of hopping conductivity in a three-dimensional system was derived by Mott as[13]

$$\sigma_{hc}(T) = \sigma_0 \exp(-A/T)^{1/4}, \tag{1}$$

where σ_0 is a material constant and A corresponds to a characteristic temperature of the system. As shown in Fig. 4, the natural logarithm of the resistivity at zero field is proportional to $T^{-1/4}$ for the two compounds, indicating that the experiment data are satisfactorily fitted in terms of the VRH model [i.e., Eq. (1)] at $T \geqslant T_N$. Thus, the VRH mechanism is believed to be responsible for the transport properties at this temperature range. The different transport property at $T \leqslant T_N$ is due to the scattering of spins. In impurity conduction in doped crystalline semiconductors with hopping conductivity, a large positive magnetoresistance can occur owing to the contraction of the overlap of the localized state wave functions in a magnetic field and, thus, an increase of the needed average hopping length (L_{hop}).[14,15] This means that the hopping probability between sites decreases (only charge carriers with larger L_{hop} can transport from one "island" to another island) and a positive MR is presented. In the paramagnetic state at $T > T_N$ for both compounds, the sign of MR is positive as shown in Figs. 2 and 3(a). This positive MR above T_N can be explained by shrinkage of the orbits in a magnetic field. Its small positive MR ratio is due to the

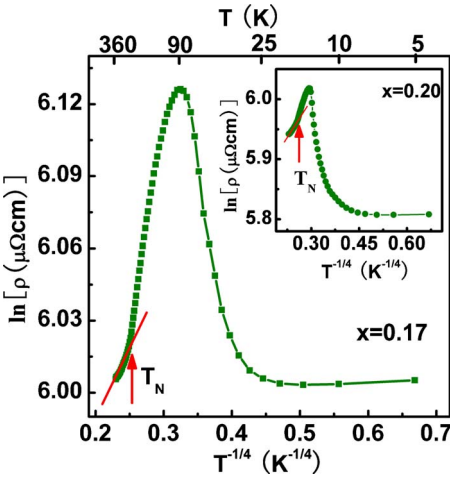

FIG. 4. (Color online) The natural logarithm of resistivity as a function of $T^{-1/4}$ for $(Mn_{0.83}Ge_{0.17})_{3.25}Ge$ and inset is that for $(Mn_{0.8}Ge_{0.2})_{3.25}Ge$ at zero field.

quantum interference caused by the increase of the temperature.

For the positive MR appearing below T_t, its mechanism is more complicated. One has to compare the MR ratio with magnetization (or spin configuration) for the two compounds below T_t. The spin structure of $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ below T_t is a collinear antiferromagnetic configuration with positive MR that monotonously increases with decreasing temperature, while that of $(Mn_{0.8}Fe_{0.2})_{3.25}Ge$ turns to a ferromagnetic configuration below T_C, accompanied with the drop of the positive MR ratio. Thus, the anomalous MR effect is spin dependent. There are three factors (spin dependent) which contribute to the magnetoresistance: spin-dependent scattering, spin-orbit effect, and spin splitting (Zeeman effect). These effects interact with each other and it is difficult to distinguish them. Spin-dependent scattering and superzone gap can account for the negative MR appearing in superlattice[16–18] and bulk compounds with magnetic transitions between ferromagnetic (or ferrimagnetic) and antiferromagnetic [such as $NdCu_2$,[19] UNiGa,[20] FeRh,[21] $Mn_2Sb_{1-x}Sn_x$,[22] and $Mn_3Zn_ySn_{1-y}C$ (Ref. 23)], in which the maximum MR usually appears around the magnetic transition temperatures. A large positive MR was reported in $RE_2Ni_3Si_5$ (Ref. 24) and $LaMn_2Ge_2$ (Ref. 25), with a naturally occurring layered structure containing sheets of rare-earth atoms, which was speculated to originate from the multiple reflections of carriers from the interface before scattering, thereby increasing the sensitivity of resistance to momentum loss upon reflection.[26,27] This mechanism cannot explain the positive MR appearing in the ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ compounds because its structure cannot be considered a layered one because there are only two crystallographic sites: $z=1/4$ and $3/4$.

The magnetoresistance of a weakly disordered electron gas arising from spin splitting of conduction-electron energies was calculated and found to be positive.[28,29] According to their calculation, MR scales as H^2 for small fields and as $\sqrt{H}$ for large fields. According to the data for the present two compounds, the spin splitting cannot explain its positive MR.

Spin-orbit (SO) scattering from paramagnetic impurities in nonmagnetic metals is known to have a significant influence on the quantum corrections. It can reverse the sign of the localization correction (so-called weak antilocalization effect), which results in a positive magnetoresistance at weak magnetic fields.[30–32] The situation in ferromagnetic metals, however, is significantly different. Theoretical study showed that the processes leading to weak antilocalization in nonmagnetic systems are totally suppressed in ferromagnets. Therefore, the presence of SO interaction results in only a negative magnetoresistance.[33]

Yamada and Tanaka[34] showed theoretically that a positive magnetoresistance can arise in a collinear antiferromagnetic metal with localized magnetic moments. The magnetic field increases (or suppresses) the spin fluctuation of the magnetic sublattice parallel (or antiparallel) to the field. The positive MR results from a competition between the suppression and the enhancement of the spin fluctuations. In rare-earth and transition metal (RT) compounds, such as UCu_2Ge_2 (Ref. 35) and $CeFe_2$, (Ref. 36) the change of MR from negative to positive was attributed to the magnetic transition from ferromagnetism to antiferromagnetism in these systems. Therefore, the theoretical calculation seems suitable to explain the positive MR obtained below T_t with collinear antiferromagnetic configuration for our compounds.

The magnetic-field dependence of MR for $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ at temperatures below T_t could be described as

$$\Delta\rho/\rho = \alpha H + \beta H^2, \tag{2}$$

where α and β are constants dependent on temperature. At temperatures far below T_t, MR implies a stronger quadratic linear dependence on H ($\beta \gg \alpha$), while a linear dependence on H at temperatures near T_t. For RT compounds mentioned above, positive MR showed a stronger linear dependence on H for all positive MR, and the same phenomenon was also observed for α-Mn below the AF transition temperature.[37] The normal solution of the Boltzmann equation for magnetoresistance yields a dependence on H^2, although scattering off the lattice and/or spin fluctuations can produce $\Delta\rho/\rho \propto H$. Semiclassically, the orbital magnetoresistance is controlled by the product of the cyclotron frequency (ω_c) and the scattering time (t), with a positive, quadratic magnetoresistance expected to saturate for $\omega_c t < 1$.[38] In this sense, the quadratic field dependence of magnetoresistance is related to nonmagnetic metals and alloys, and the linear field dependence is related to magnetism. However, for our $(Mn_{0.83}Fe_{0.17})_{3.25}Ge$ compound with antiferromagnetic configuration, the quadratic field dependence of MR is strange. As we mentioned above, the unit cell volume dropped by about 2% at and/or below T_t for $(Mn_{0.75}Fe_{0.15})_{3.1}Ge$.[8] The decrease of the unit cell may lead to the change of the surface of the Fermi surface. The quadratic H dependence is expected due to Fermi surface effects.[35] The linear dependence of positive MR ratio on H near and/or below T_t is due to disorder and inhomogeneity[39] near the magnetic transition, which can be seen in the steplike magnetization curve in Ref. 10.

IV. SUMMARY

The magnetic, transport, and magnetotransport properties of the ε-$(Mn_{1-x}Fe_x)_{3.25}Ge$ compounds with $x=0.17$ and 0.2 have been investigated systematically. Anomalous MR effect was observed, namely, a substantial positive MR below T_t (magnetic transition from triangular to collinear antiferromagnetic configuration), a small negative MR between T_t and T_N, and a positive MR above T_N. The origin of the anomalous MR effect has been discussed in terms of the shrinkage of the orbits, the spin fluctuations, the spin configurations and the magnetic transitions.

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