Large magnetoresistance and metamagnetic transition in PrGa

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The magnetic phase transition and the magnetoresistance (MR) in bulk intermetallic PrGa compound are investigated experimentally. Two successive magnetic transitions, ferromagnetic (FM)-antiferromagnetic (AFM) transition and AFM-paramagnetic transition, are observed at $T_1 = 28$ K and $T_2 = 36$ K, respectively. It is found that the PrGa compound exhibits a field-induced metamagnetic transition from AFM to FM state and a considerable change in lattice constants in a temperature range of 28-36 K. Accompanied with the AFM-FM transition, a negative MR occurs, and the maximal MR values are ~30% and ~34% at 28 K under the fields of 1 T and 5 T, respectively. © *2011 American Institute of Physics*. [doi:10.1063/1.3641879]

In the bulk intermetallic materials, giant magnetoresistance (GMR) effect usually occurs at a magnetic-field-induced magnetic transition from antiferromagnetic (AFM) (or paramagnetic (PM)) to ferromagnetic (FM) state, which is frequently accompanied by a crystallographic transformation or considerable volume change. Typical examples of this class of materials are natural layered $SmMn_2Ge_2$ (Ref. 1) and FeRh alloy[2] and bulk intermetallic compound with magnetostructural transition $Gd_5Si_{1.8}Ge_{2.2}$.[3] As is known, the mechanism for GMR behavior is dependent on the nature of the crystallographic/magnetic structure and phase transition of the material. Various theoretical interpretations have been proposed for the origin of different GMR behaviors. The large magnetoresistance (MR) effect in FeRh, an AFM alloy with layered structure, is believed to correlate with both the spin-flip transition of Fe magnetic moment and the onset of Rh magnetism.[4] As for $Gd_5Si_{1.8}Ge_{2.2}$,[3] it is presumed that the occurrence of GMR effect may be due to the variation in density of states at the Fermi level caused by the crystal symmetry change. Evidently, extensive studies of the GMR effect in a variety of materials are worthwhile both for the fundamental physics and for the technological application.

The magnetic properties of *R*Ga (*R* = rare earth) compounds have been systematically investigated by means of neutron diffraction, Mössbauer spectroscopic, and magnetic measurements.[5–7] However, there exist some discrepancies among the reported values of the magnetic ordering temperatures. The FM–PM phase transition temperature of PrGa was reported to be 32.5 K by Nesterov *et al.*[6] and 36 K by Delyagin *et al.*[7] Our experimental results show that PrGa have two ordered magnetic states in low magnetic fields. It orders ferromagnetically below 28 K and experiences a magnetic transition to AFM state when being heated. In addition, it is also found that the magnetic field can induce an AFM-FM transition, which is accompanied by a considerable change in lattice constant and a large MR.

The polycrystalline PrGa compound was prepared by arc melting the constituent elements with a purity of better than 99.9% in argon atmosphere. The ingot was annealed in an evacuated sealed quartz tube at 800 °C for 1 week to improve homogeneity and crystallinity. The room-temperature x-ray powder diffraction pattern confirmed the single-phase nature of the compound, crystallizing in the orthorhombic CrB-type structure (space group Cmcm). Heat capacity, magnetization, electrical resistance, and thermal expansion were measured by employing a physical property measurement system (PPMS) from Quantum Design.

The values of heat capacity (C_p) as a function of temperature were measured in various magnetic fields as shown in Fig. 1(a). Double peaks are observed in zero field at two successive temperatures, $T_1 = 28$ K and $T_2 = 36$ K, respectively, suggesting the occurrence of two phase transitions. The position of the low-temperature peak, which strongly depends on the applied magnetic field, shifts rapidly toward higher temperature with field increasing. However, the position of the high-temperature peak remains almost unchanged. When a field larger than 1 T is applied, only a single peak is observed on the C_p-T curve. To get a better insight into the magnetic nature of two transitions, the temperature and the magnetic field dependences of the magnetization (M) of PrGa are investigated in detail. Figure 1(b) shows the temperature dependences of zero-field-cooling (ZFC) and field-cooling (FC) magnetizations under a magnetic field of 0.01 T. The behaviors of M–T curve for PrGa are very similar to those observed in Tb_3Co, where the FM–AFM and AFM–PM transitions occur in sequence with temperature increasing.[8] For PrGa, the abrupt change in magnetization around T_1 should coincide with the magnetic transition from FM to AFM state. The transition temperature T_1, defined by the minimum value of dM/dT, is determined to be 28 K in good agreement with the value obtained by the position of the low-temperature peak on the C_p-T curve. The small cusp at T_2 as shown in the inset of Fig. 1(b), corresponding to the high-temperature peak on the heat capacity curve, is believed to be an

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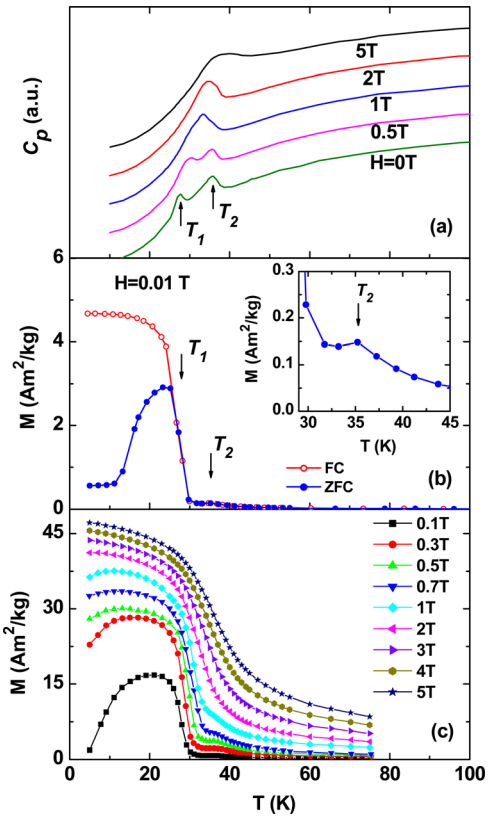

FIG. 1. (Color online) (a) Heat capacity as a function of temperature for PrGa in various fields. (b) Temperature dependences of the ZFC and the FC magnetizations of PrGa in a magnetic field of 0.01 T. (c) Temperature dependences of the magnetizations of PrGa at different magnetic fields.

indication of an AFM-PM phase transition. The temperature dependences of magnetization in various magnetic fields are plotted in Fig. 1(c). It is noticeable that the magnetization in AFM region, namely in the temperature range between T_1 and T_2, increases greatly with the increase of magnetic field, revealing the occurrence of a field-induced AFM-FM transition. As shown in Fig. 1(c), the AFM state completely transforms into the FM state when a magnetic field higher than 1 T is applied, and a field-induced FM-PM phase transition occurs, which is pushed toward higher temperature by applied magnetic field.

The magnetization isotherms of PrGa were measured. Figure 2 displays a set of selected *M–H* curves for the sake of clarity. As is shown, the curves show typical FM and PM behavior at temperatures below T_1 and well above T_2, respectively. The magnetization is observed to increase linearly with the increase of the magnetic field between T_1 and T_2 in a low field region, indicating the existence of AFM state. Further increase of the field makes the magnetization curve deviate from such a linear relationship, indicating the onset of a field-induced metamagnetic transition from AFM to FM state. The critical magnetic field for this metamagnetic transition, determined from the maximum of the dM/dH curve, is found to increase from 0.35 T at 29 K to 0.9 T at 35 K, indicating that the AFM interaction in PrGa between T_1 and T_2 is relatively weak and can be easily overcome by magnetic field.

Figure 3(a) shows the electrical resistivity (ρ) as a function of temperature measured in magnetic field of 0 T, 2 T, and 5 T. In zero field, the resistivity exhibits a temperature dependence typically observed in metallic systems, but it

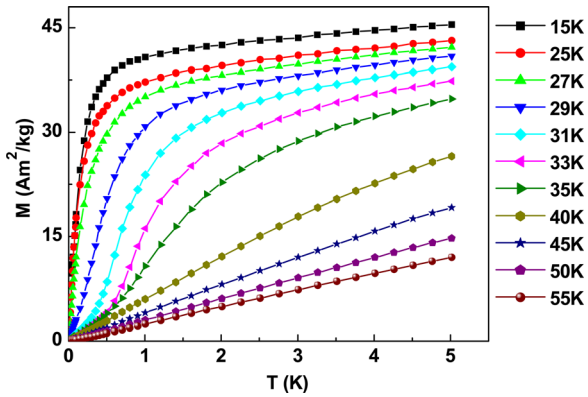

FIG. 2. (Color online) Magnetization isotherms of PrGa in a temperature range of 15–55 K under the magnetic fields up to 5 T.

slightly increases below T_2 as the temperature decreases and shows a maximum as often observed in AFM materials[4,9,10] before it decreases sharply below 28 K. Such variations in the value of ρ lead to a large positive peak and a minimum on the $d\rho/dT$ curve at $T_1 = 28$ K and $T_2 = 36$ K, respectively, as shown in the inset of Fig. 3(a), which should correspond to the FM-AFM and AFM-PM transitions, respectively. The small increase in resistivity below T_2 comes from the reduction of the effective number of conduction electrons caused by the Fermi level gapping due to a new magnetic periodicity of long-range AFM spin ordering, and the dramatic drop of resistivity below T_1 should result from the temperature-induced transition from high-resistivity AFM to low-resistivity FM phase. It can be also seen from Fig. 3(a) that when the magnetic field of 2 T and 5 T are applied, there is a negligible change in ρ above 45 K. However, as the

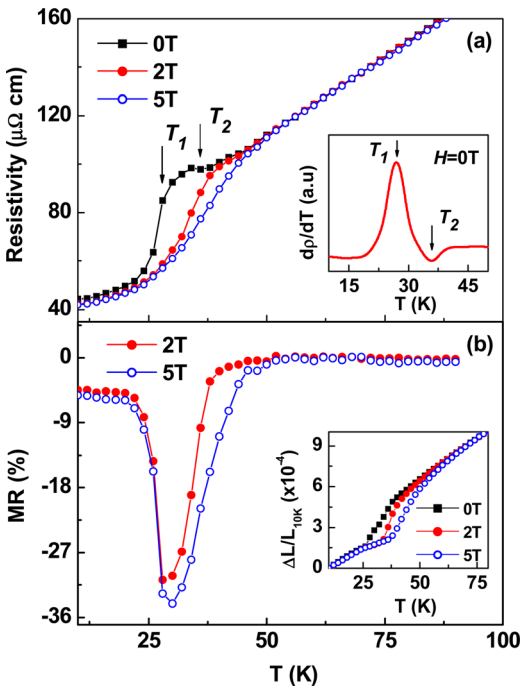

FIG. 3. (Color online) (a) Temperature dependence of electrical resistivity for PrGa under the field of 0 T, 2 T, and 5 T. The inset shows the temperature dependence of $d\rho/dT$ in zero field. (b) Temperature dependence of magnetoresistance under the field of 2 T and 5 T. The inset shows the thermal expansion of PrGa under the field of 0 T, 2 T, and 5 T.

temperature is lowered toward T_2, the value of ρ becomes small compared with that in zero field, strongly depending on the applied field strength. The maximum on the ρ–T curve found between T_1 and T_2 in the zero-field, which is related to the AFM phase, is totally invisible due to the AFM-FM metamagentic transition.

The temperature dependent MR is plotted in Fig. 3(b) in 2 and 5 T field. The ratio $[\rho(H,T) - \rho(0,T)]/\rho(0,T)$ is used to represent the MR. One can see that the PrGa compound exhibits a negative MR effect in the entire temperature range studied. In low temperature range, the magnitude of MR is rather small, which is as expected for the normal FM state, whereas it is enhanced significantly between T_1 and T_2. Figure 4 shows the magnetic field dependence of the MR in a temperature range of 15-50 K. It is observed that the MR drops significantly with the increase of field when the magnetic field smaller than ~1 T around T_1, then retains a large value with the further increase of field. The result displays a clear signature of the field-induced metamagnetic transition from high-resistance AFM to low-resistance FM state. It is worth noting that the MR value is nearly as large as 30% at 28 K when a relatively small field (1 T) is applied, which is beneficial for the potential technology applications of the present sample. For the PrGa compound, the values of MR are found to be ~34% and ~35% at 28 and 30 K in a magnetic field of 5 T, respectively. The maximal MR in PrGa is comparable to or much larger than those of some best known intermetallic compounds with field-induced metamagnetic transitions, such as $SmMn_2Ge_2$ (8%),[1] $Ce(FeRu)_2$ (20%),[9] Gd_2In (29%),[10] $Gd_5(Si_2Ge_2)$ (26%),[11] and $Gd_5(Si_{1.8}Ge_{2.2})$ (20%).[3] Theoretical study based on the independent scattering mechanism shows that in metallic systems, the difference in resistivity owing to the magnetic structure modification from AFM to FM configuration correlates well with the exchange interactions acting on the conduction electrons and the magnons.[4,12] Generally, this difference between regular-rare-earth intermetallic compounds is rather modest because the coupling of the conduction-electron spin and the ionic spin moment is weak. Therefore, the large difference in resistivity between AFM and FM states in the present sample cannot be explained solely by considering the contribution of the magnetic structure modification to the change in electron-magnon scattering. Some other effects should be also taken into account. It is found that the transformation of the magnetic configuration is frequently accompanied by a crystallographic structure transition or a noticeable change of the lattice volume. Both magnetic and structural transitions (or volume change) can lead to the changes in the electron-magnon scattering and the conduction electron concentration, each of which plays a role in the change in the electrical resistivity. To well comprehend the large MR effect in PrGa, we measure the thermal expansion data ($\Delta L/L_{(10K)}$) under the fields of 0, 2, and 5 T by means of strain gauge method (Ref. 13) as shown in the inset of Fig. 3(b). Results show that the $\Delta L/L_{(10K)} - T$ curve in zero field increases almost linearly with the increase temperature below T_1. However, it deviates from this linear relationship around T_1 and increases sharply with the increase of temperature, revealing the occurrence of the abrupt thermal expansion, namely great change in lattice constant, during the transition from FM to AFM state. In addition, the $\Delta L/L_{(10K)}$ value is lowered considerably in the AFM regime, respectively, at applied magnetic fields of 2 T and 5 T, implying that the great change in lattice volume also takes place at the field-induced AFM-FM transition. Based on the magnetic and strain measurements, it is believed that the noticeable MR values in PrGa might be attributed to the great changes in electron-magnon scattering and the conduction electron concentration caused by both the magnetic structure transformation from AFM to FM configuration and the associated great volume change.

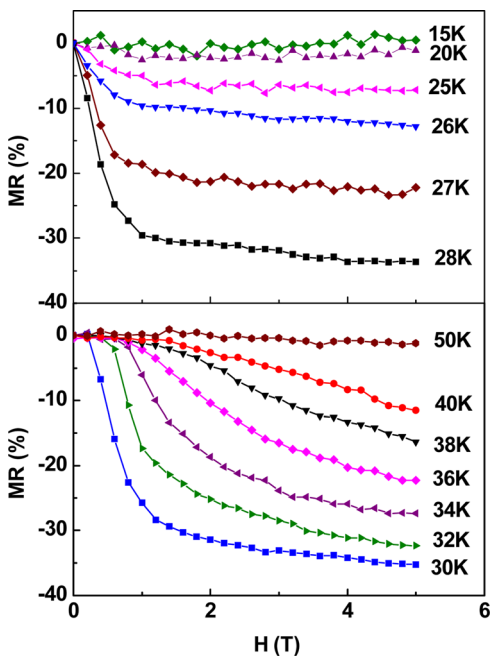

FIG. 4. (Color online) Magnetic field dependence of magnetoresistance for PrGa in a temperature range of 15–50 K.

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