Giant magnetoresistance in Ce_2Fe_{17}

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Using a single-phase sample of Ce_2Fe_{17} with the rhombohedral Th_2Zn_{17}-type structure, we measured the magnetization, resistivity, and specific heat under various magnetic fields up to 5.5 T. Below $T_c = 125$ K, giant magnetoresistance (GMR) is induced by the metamagnetic transition from the antiferromagnetic to ferromagnetic states. The value of $\Delta\rho/\rho(\cong \rho_{AF} - \rho_F)/\rho_{AF})$ reaches 0.85 at 4.2 K, while the value $\gamma (\cong C/T$ at 0 K) of 351 mJ/K^2/mole in zero field only decreases $\sim$20% by the metamagnetic transition. These results suggest that GMR is due to the closing of a superzone gap formed by antiferromagnetic ordering of Fe spins, while the large γ value is due to valence fluctuations of the Ce-4f electrons, weakly dependent on magnetic field. [S0163-1829(97)03046-4]

I. INTRODUCTION

Since the discovery of high-T_c superconductors, there has been interest in some anomalous transport properties in highly correlated electron systems, in which spins and charges interplay.

One of the typical examples is an insulator-metal transition phenomenon depending on band filling, temperature, and magnetic fields for prototype double exchange ferromagnets, $La_{1-x}Sr_xMnO_3$.[1] Around T_c, a colossal magnetoresistance, caused by suppression of spin fluctuations, was observed. For the uranium compound UNiGa, it has been found that the electrical resistivity displays a dramatic decrease due to the metamagnetic transition from the antiferromagnetic to ferromagnetic states.[2] In UNiGa the magnetoresistance, defined as $\Delta\rho/\rho \cong (\rho_{AF} - \rho_F)/\rho_{AF}$, reaches 0.90 at 4.2 K along the c axis of the ZrNiAl-type hexagonal layered structure. We called such an anomalously large magnetic field effect of resistivity ''giant magnetoresistance'' (GMR). This word has first been used for huge magnetoresistance in magnetic superlattices.[3] Furthermore, in reentrant ferromagnetic $SmMn_2Ge_2$ with the $ThCr_2Si_2$-type layered structure,[4] GMR has been observed around 125 K, where a metamagnetic transition from an antiferromagnetic to induced ferromagnetic states occurs.

However, no anomalous transport properties have yet been reported in the rare-earth iron intermetallics, as far as we know. Recently, we found GMR at low temperatures in Ce_2Fe_{17} crystallized in the rhombohedral Th_2Zn_{17}-type structure. In this paper, we will present our results of the GMR effect in Ce_2Fe_{17}. The binary Ce-Fe phase diagram indicates a difficulty of producing single-phase Ce_2Fe_{17}.[5] Hence, nobody has made systematic studies on the physical properties

for Ce_2Fe_{17}. Only a few works have been independently studied by measurements of magnetization,[6,7] Mössbauer effect,[8] specific heat,[9] x-ray absorption,[10] and neutron diffraction.[11]

II. EXPERIMENTS

The initial compound was prepared by arc melting stoichiometric amounts of the constituent metals Ce and Fe with a purity of 99.99% under Ar flow atmosphere. The ingot was turned over and remelted several times to ensure homogeneity. After wrapping it in Ta foil the ingot was annealed at $\sim$1310 K for one week in a vacuum tube together with a small piece of pure Ce, and finally it was quenched into water.

The x-ray-diffraction profile of the sample obtained is shown in Fig. 1, together with a profile calculated using RIETAN-94.[12] It is clear that the sample is a single-phase rhombohedral Th_2Zn_{17}-type sample and no trace of α-Fe or the hexagonal Th_2Ni_{17}-type phase is visible in the x-ray-diffraction pattern. We tried many times to make more single-phase samples, but they were always contaminated by impurity phases, either α-Fe, Ce oxides, or the hexagonal Th_2Ni_{17}-type phase. The results indicate that the condition to obtain single-phase Ce_2Fe_{17} is quite severely dependent on the annealing temperature and time.

Magnetization measurements were performed using a superconducting quantum interference device magnetometer in magnetic fields up to 5.5 T in temperature ranging from 5 to 300 K. Resistivity measurements were performed using a standard four-probe technique in fields up to 4 T in the temperature range from 4.2 to 300 K, with the current direction perpendicular to the field direction. Specific-heat measure-
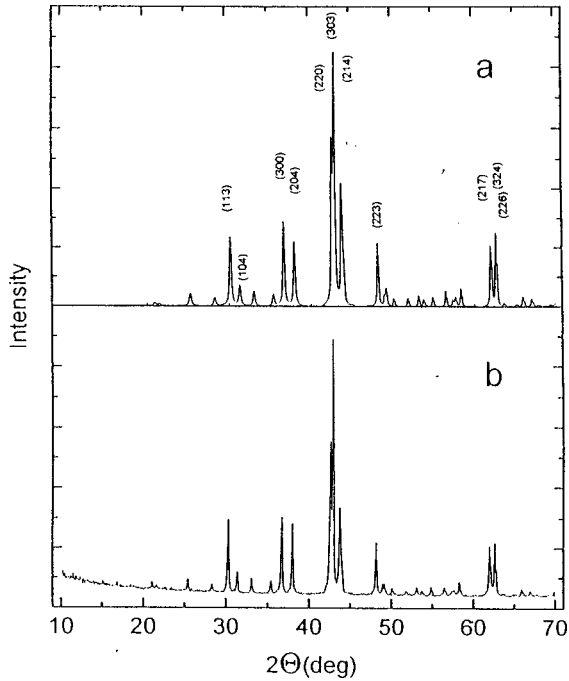
FIG. 1. Cu K-α x-ray-diffraction profiles of Ce_2Fe_{17} simulated by RIETAN (a) and obtained in this work (b).

ments were performed in fields up to 2.5 T from 2.5 to 8.3 K using a standard adiabatic calorimeter with a mechanical heat switch.

III. RESULTS AND DISCUSSION

The magnetic-field dependence of magnetization at various typical temperatures is shown in Fig. 2. At 5 K, the magnetization first increases linearly with increasing field, showing no ferromagnetic component, and then a metamagnetic transition occurs around 0.75 T. This metamagnetic transition is accompanied with a hysteresis of about 0.2 T at 5 K, which decreases with increasing temperature and disappears above 125 K. Between 65 and 125 K, the metamag-
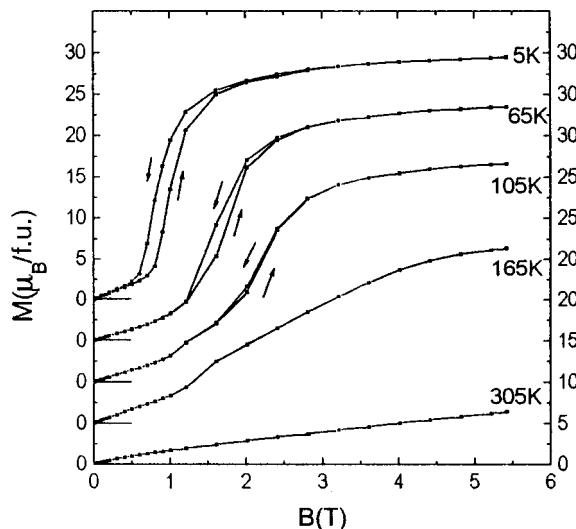

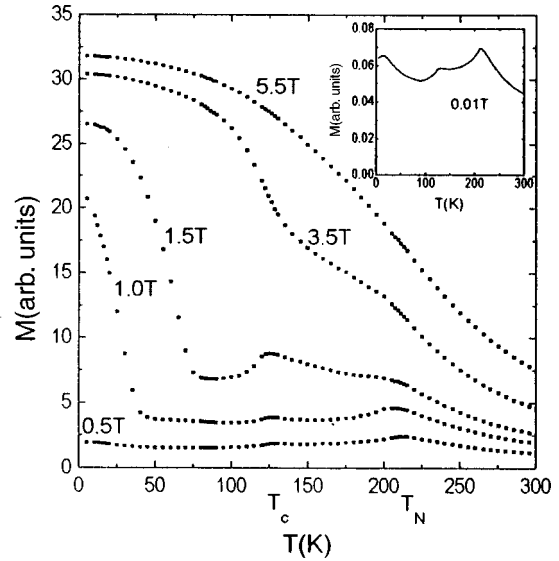FIG. 2. Magnetization vs magnetic-field curve of Ce_2Fe_{17} at various typical temperatures.

FIG. 3. Magnetization as a function of temperature for Ce_2Fe_{17} in various magnetic fields.

netism is preceded by a distinct change in the slope of the linear field dependence of the magnetization. The magnetization at 165 K shows no metamagnetic transition at lower magnetic field even though a small s-shaped feature can be distinguished in the M-B curve. Around 1.5 T, the curvature of the M-B curve suddenly changes, after which an intermediate phase appears. With further increasing magnetic field, the magnetization begins to saturate above 4 T, indicating induced ferromagnetism. A similar M-B curve was observed between 125 K and $T_N = 215$ K. Until now, the magnetization of Ce_2Fe_{17} has been measured by two different authors.[6,7] Both results indicate a small but finite ferromagnetic component in the sample, which might be due to a small segregation of an α-Fe phase. So, we believe that our sample is the best among the previous samples that have been reported so far. Figure 3 shows the magnetization as a function of temperature under various magnetic fields. The magnetization at 0.01 T is also shown in the inset. Two sharp peaks occur at 125 and 215 K, respectively. These were previously reported as a critical temperature at which the magnetic structure changes from helical to fan structure and the Néel temperature, respectively. In low field, there is also a broad maximum around 15 K, like in a spin glass. However, since this easily disappears in higher fields, it seems likely that it is not intrinsic. In all fields up to 5.5 T, a critical temperature T_c is observed at 125 K with no magnetic field dependence. On the contrary, the Néel temperature T_N slightly decreases with increasing magnetic field.

From the results obtained above, we can deduce the magnetic phase diagram that is shown in Fig. 4. Givord and Lemaire[11] have claimed that the AFI phase is a fan structure, having a ferromagnetic component, and AFII is a helix with a wave vector of 0.037 A^{-1} along the c axis. The IM phases I and II indicate intermediate phases without any ferromagnetic components different from AFI and AFII, respectively. However, it was clarified in this work, that the AFI phase is not a fan structure because this phase does not have any ferromagnetic component. The IF phases are induced ferromagnetic phases, in which the average Fe moment is
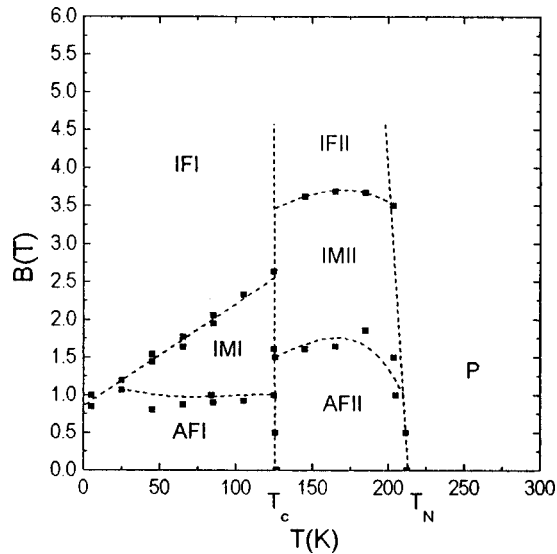
FIG. 4. Magnetic phase diagram of Ce_2Fe_{17} deduced from magnetic measurements.

FIG. 6. Resistivity as a function of temperature for Ce_2Fe_{17} under a zero field and 3 T.

$\sim 1.70\mu_B$ at 5 K, assuming that Ce is nonmagnetic in a mixed-valence state.

In Fig. 5, we show the relative change of resistivity $\rho(B)/\rho(0)$ as a function of magnetic field at 4.2 K, together with the magnetization curve at 5 K. The virgin curve is also displayed, in which the resistivity ratio $\rho(B)/\rho(0)$ slightly increases from about 0.5 T and after showing a maximum at 0.7 T, $\rho(B)/\rho(0)$ quickly decreases with increasing magnetization. Here it is to be noted that the small increase at about 0.5 T is probably caused by the formation of microcracks due to strong distortion induced by the metamagnetic transition. After taking the virgin curve, our sample was reasonably stabilized and reproducible data could be obtained. As is evident from Fig. 5, almost the same hysteresis was observable in the $\rho(B)/\rho(0)$ vs B curve as in the magnetization vs field curve. After the metamagnetic transition occurs from the antiferromagnetic AFI to the induced ferromagnetic IFI state, the value of $\Delta\rho/\rho=(\rho(0)-\rho(B)/\rho(0))$ reaches 0.85
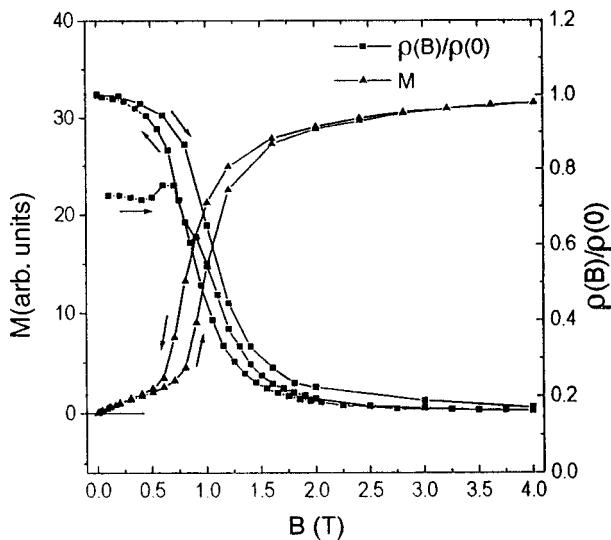
at $H=4$ T. Because of the microcracks occurring around T_c in our sample during cooling down or heating up, it was difficult to accurately measure the temperature dependence of the resistivity. However, we were able to produce the ρ vs T curve in zero field using overlapping datasets for the whole temperature range, but were not able to produce it in 3 T above 125 K. Probably also due to induced microcracks in the sample, the absolute value of the resistivity could not be accurately determined in this work. It is noteworthy that the lowest (most correct) value at 300 K was ~ 400 $\mu\Omega$ cm. As is seen in Fig. 6, the shape of the ρ vs T curve below T_c in zero field is reminiscent of gap formation due to development of a spin-density wave like in Cr-Mn alloy.[13] This simply suggests that a superzone gap is opened below T_c in Ce_2Fe_{17}. In the field of 3 T, where the induced ferromagnetism is stabilized, the resistivity becomes quite small compared to the zero-field value, and linearly increases with in-

FIG. 5. Resistivity (normalized) and magnetization as a function of magnetic field for Ce_2Fe_{17}.

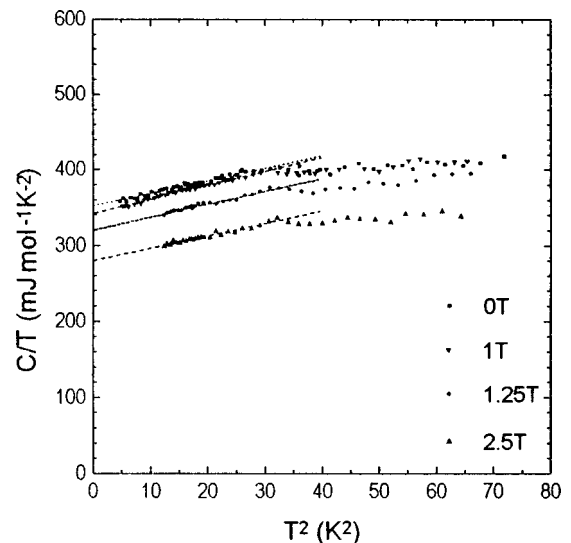FIG. 7. Specific heat of Ce_2Fe_{17} in various magnetic fields. The straight lines denote the linear extrapolations to obtain γ ($=C/T$ at 0 K).

creasing temperature. Therefore, GMR in Ce_2Fe_{17} seems to be induced as a result of closing a superzone gap by magnetic field.

As another possibility for the origin of GMR, we consider it to be due to remaining spin fluctuations of magnetic spins in the Fe sublattices even at 4.2 K. If this is the origin of GMR, the T linear term of the specific heat should be strongly reduced by suppression of spin fluctuations due to induced ferromagnetism. In Fig. 7, we demonstrate the results of the low-temperature specific-heat measurements under magnetic fields. By extrapolating the T linear term of specific heat C/T vs T^2 curve to $T=0$, we obtained the γ value of 351 mJ/K^2/mole in zero field, with the definition $\gamma = C/T$ at 0 K. This value is significantly lower than the previously reported value of 560 mJ/mole/K^2.[9] This might be due to the sample quality. In magnetic fields, the γ value is suppressed, leading to a reduction of $\sim$20% in 2.5 T in the induced ferromagnetic state. Such a weak suppression of the γ value in magnetic field denies the possibility for the ap-

pearance of GMR to be due to the fluctuation of the Fe spins at low temperatures. So, we can conclude that GMR is mainly originated in the development of a superzone gap due to a specific ordering of Fe spins below T_c. The suppression of γ might be due to a change in valence fluctuations of the Ce $4f$ electrons.

In order to clarify the origin of GMR in Ce_2Fe_{17}, we are now planning to perform the neutron-diffraction studies using a single-phase sample.

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