Magnetoresistance and magnetization studies of the Laves-phase compound $Ce(Fe_{0.92}Al_{0.08})_2$

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(Received 16 May 1994)

A detailed study of magnetoresistance and magnetization is reported on the Laves-phase compound $Ce(Fe_{0.92}Al_{0.08})_2$ in the temperature range of 4.4 to 250 K and in magnetic fields up to 45 kOe. The measurements indicate a paramagnetic to ferromagnetic transition at 170 K and rule out the possibility of a spin-glass transition as had been earlier reported in some studies. The magnetoresistance measurements clearly indicate that the compound has an antiferromagnetic state below 100 K as suggested by earlier neutron-scattering measurements. The antiferromagnetic state is found to undergo a metamagnetic transition which could be observed in the present study above 60 K.

I. INTRODUCTION

Among the Laves-phase compounds, $CeFe_2$ has been extensively studied and is found to show ferromagnetism below 230 K.[1] The substitution of Fe by even small concentrations of Al, Co or Ru destabilizes the ferromagnetism as is observed from studies on the pseudobinary intermetallics $Ce(Fe,M)_2$ (M=Al, Co, Ru).[2,3] In these compounds, one observes a second transition to an antiferromagnetic (AF) state from a ferromagnetic (FM) state as the temperature is lowered. In $Ce(Fe,Al)_2$, the instability of the FM phase is observed even with 2% Al substitution at the Fe site. For Al concentrations below 5%, ac susceptibility and magnetization results resemble reentrant spin glasses. However, resistivity results suggest that the low temperature phase is antiferromagnetic in nature.[3] For Al concentrations above 5%, bulk property measurements indicate the presence of a para- to antiferromagnetic transition.[3] The existence of short-range or inhomogeneous magnetic ordering in the so-called paramagnetic phase, however, cannot be ruled out. Most of these studies have been carried out for Al concentrations less than 10% where a single crystallographic pure $C15$ phase is found. In another study of magnetization and resistivity on compounds up to 15% Al substitution, the authors emphasize that the presence of Al destroys ferromagnetic order and leads to a reentrant spin-glass or spin-glass phase.[4] However, neutron-scattering studies on the above pseudobinary compounds have confirmed the presence of an antiferromagnetic ordering at low temperatures.[5] In $Ce(Fe,Al)_2$, the transition from the FM to AF state is a gradual one, passing through a region of coexistence of FM and AF moments as has been shown from the ac susceptibility,[3] and magnetization studies[6] of this system up to 20% Al concentration. The possibility of a spin-canted phase in the coexistence regime has been predicted and is also supported by neutron diffraction,[5] hyperfine field,[7] and nonlinear ac susceptibility measurements[8] on these systems.

Some of the Laves-phase compounds, such as RCo_2 (R=Y, Lu, etc.) show a metamagnetic transition at very high fields of the order of a few 100 kOe.[9] They undergo a field-induced transition from a paramagnetic state to a ferromagnetic state. The present study as well as some earlier studies[6] show that the AF state of the $Ce(Fe,Al)_2$ compound undergoes a metamagnetic transition to a ferromagnetic state at much lower fields than those for RCo_2 compounds. There have been attempts to obtain theoretical phase diagrams for an itinerant electron system having competing ferromagnetic and antiferromagnetic interactions with and without magnetic anisotropy.[10,11] These studies predict a number of temperature- and field-dependent magnetic states which are relevant to our present study.

In the present paper, we report the magnetoresistance ($\Delta\rho/\rho$) and magnetization measurements on the Laves-phase compound $Ce(Fe_{0.92}Al_{0.08})_2$ in the temperature range 4.4–200 K up to fields of 45 kOe. Magnetoresistance measurements in this system reveal features ascribed to metamagnetic behavior in the low temperature AF state.

II. EXPERIMENTAL DETAILS

The sample used for the present study has been used earlier for ac susceptibility and resistivity measurements.[3] Details of the sample preparation and

characterization can be found in that paper. The sample used in the present measurements was a square cross-section rod of dimension $20 \times 1 \times 1$ mm^3. As has been described earlier,[3] a very small amount of magnetic impurity (which evades detection in the conventional method, e.g., x-ray diffraction) remained in the sample and made its presence felt in the low-field studies. However, in our present study, which is mostly confined in the high-field regime, this small amount of impurity will not influence the results in any appreciable manner.

The longitudinal magnetoresistance (MR) measurements were carried out in the temperature range 4.4 to 250 K up to fields of 45 kOe generated by a home-built superconducting magnet. The magnetoresistance was measured using standard four-probe dc technique. The electrical contacts to the sample were made with indium solder using ultrasonic soldering. The temperature of the sample was controlled and monitored by a Lake shore carbon glass sensor (in magnetic field) up to 60 K and by a silicon diode sensor beyond 60 K employing a Lake Shore DRC-82C temperature controller. The sample was cooled to 4.4 K in zero external magnetic field and the data were recorded by increasing the field in steps at discrete intervals up to 45 kOe. For subsequent temperatures above 4.4 K, the sample was heated to those temperatures in zero field and the data taken in a similar manner as described above.

The magnetization measurements reported here were performed using a commercial superconducting quantum interference device (SQUID) magnetometer (MPMS5, Quantum Design). The sample was mounted firmly in the sample holder with its long axis parallel to the external magnetic field. The sample position and configuration were strictly maintained during all the measurements in order to avoid any orientational effect due to anisotropy (if any). The scan length we used was 4 cm and measurements were averaged over three scans each containing 32 data points. This relatively short scan length minimizes the inhomogeneity of the magnetic field through which the sample travels. Before each measurement, special care was taken to reduce the trapped field in the superconducting magnet of the SQUID magnetometer.

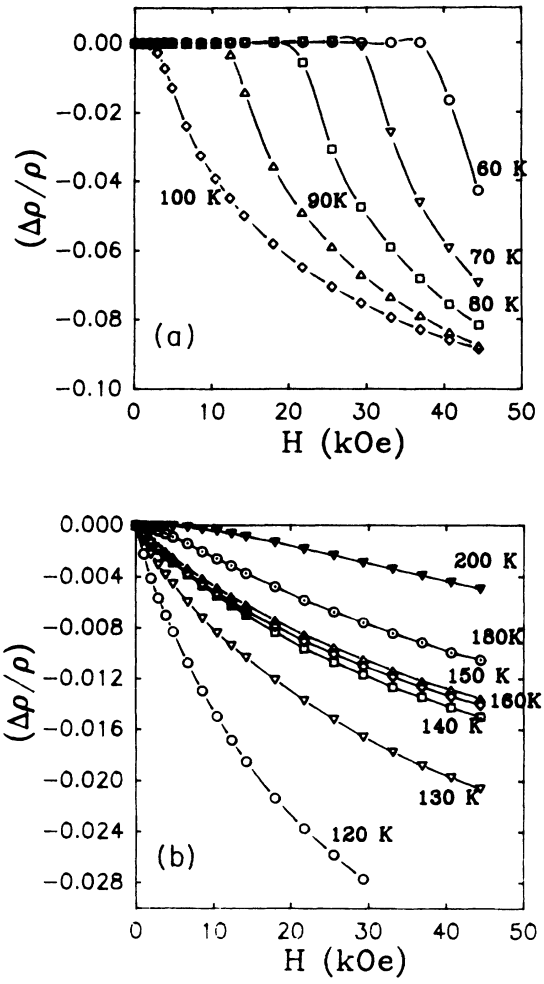

FIG. 1. (a),(b) Magnetic field (H) dependence of the magnetoresistance ($\Delta\rho/\rho$) in $Ce(Fe_{0.92}Al_{0.08})_2$ at different temperatures.

III. RESULTS AND DISCUSSION

A. Field dependence of the magnetoresistance

Figures 1(a) and 1(b) shows the magnetic field dependence of magnetoresistance ($\Delta\rho/\rho$) at different temperatures. Below 60 K, ($\Delta\rho/\rho$) is negligibly small (less than 0.1% at the maximum applied field of 45 kOe). At 60 K, the sudden development of negative magnetoresistance around 37 kOe indicates that the compound undergoes a metamagnetic transition from an antiferromagnetic to a ferromagnetic state. A similar transition in magnetoresistance has earlier been observed by us in UCu_2Ge_2.[12] The field (H_m), at which negative magnetoresistance develops, decreases with increasing temperature up to 100 K. Above the metamagnetic transition, the magnetoresistance does not saturate up to the maximum applied fields of 45 kOe. This indicates that the system does not acquire a collinear ferromagnetic state but rather goes into a canted ferromagnetic state in the range of fields studied. Between 100 K and 160 K, the field dependence of $\Delta\rho/\rho$ is similar to that observed in inhomogeneous ferromagnets such as $Au_{82}Fe_{18}$,[13] amorphous FeNiPBAl,[14] etc. At 200 K, $\Delta\rho/\rho$ tends towards a quadratic field dependence as is the case for paramagnets.[13]

The field (H) dependence of magnetization (M) at a few temperatures is shown in Fig 2. At 60 K and 80 K, we find that the magnetization rises initially and then saturates at a field between 2 and 3 kOe. Further at a higher field (depending on temperature), the magnetization starts increasing rather rapidly showing a metamagnetic transition. This transition is accompanied by the onset of marked hysteresis in the M vs H plots. The field at which magnetization shows the metamagnetic transition coincides with H_m. No saturation in magnetization

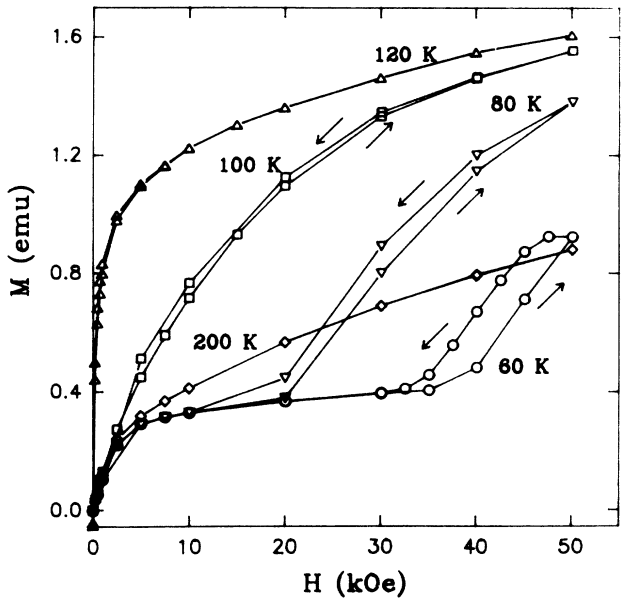

FIG. 2. Magnetic field (H) dependence of the magnetization (M) in $Ce(Fe_{0.92}Al_{0.08})_2$ at different temperatures. Arrows indicate data taken in increasing and decreasing fields.

is observed up to fields of 50 kOe which is the maximum available field in the present measurement. Since the magnetoresistance at low fields shows an antiferromagnetic behavior, the initial increase of magnetization seems to occur due to presence of an impurity phase. The magnetization behavior at 100 K shows that the metamagnetic transition field is very low, giving rise to a continuously increasing magnetization. The behavior at 120 and 200 K is similar to that of an inhomogeneous ferromagnet where the magnetization initially rises rapidly and then slowly with increasing fields.[15]

This kind of field-induced transition has been observed in the magnetization studies on some Al- and Ru-substituted systems.[6,7,16] The temperature dependence of H_m shows almost a linear decrease with increasing temperature as seen in Fig 3. From their magnetization study on Ce(Fe, 5% Al)$_2$, Nishihara *et al.*[7] showed the temperature dependence of the transition field at which magnetization saturates. This dependence is qualitatively similar to the one observed in the present study.

B. Temperature dependence of the magnetoresistance

Figure 4 shows the temperature dependence of the magnitude of the magnetoresistance ($|\Delta\rho/\rho|$) in different magnetic fields. In a given field, the magnetoresistance is negligibly small in the AF state. In the range where the metamagnetic transition is found to occur, $|\Delta\rho/\rho|$ increases rapidly with temperature. The maximum in $|\Delta\rho/\rho|$ occurs at 100 K which is independent of applied field, as can be seen in the figure. Above 100 K, $|\Delta\rho/\rho|$ drops rapidly and shows a small hump particularly at high fields, at around 170 K which is close to the temperature where the ferromagnetic moment develops at the Fe site[5] and a peak in ac susceptibility (χ_{ac}) has been observed.[3]

The neutron diffraction study on Ce(Fe,Al)$_2$ by Kennedy and Coles[5] shows that even in $CeFe_2$ there is a tendency towards antiferromagnetic ordering. Here the development of ordered moments is seen at both Ce and Fe sites and the coupling between them is found to be antiparallel. In $Ce(Fe_{1-x}Al_x)_2$ ($x < 0.08$), no magnetic moment on Ce is seen in the FM state though the low temperature state of the compound is antiferromagnetic.

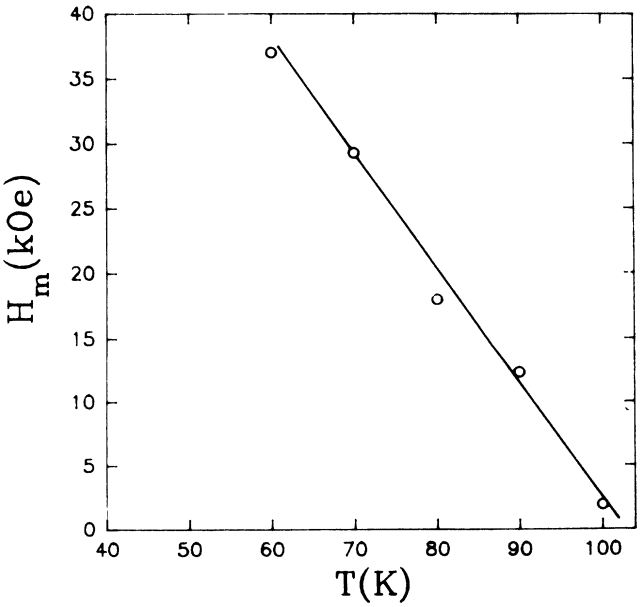

FIG. 3. Temperature variation of the field (H_m) at which the metamagnetic transition occurs.

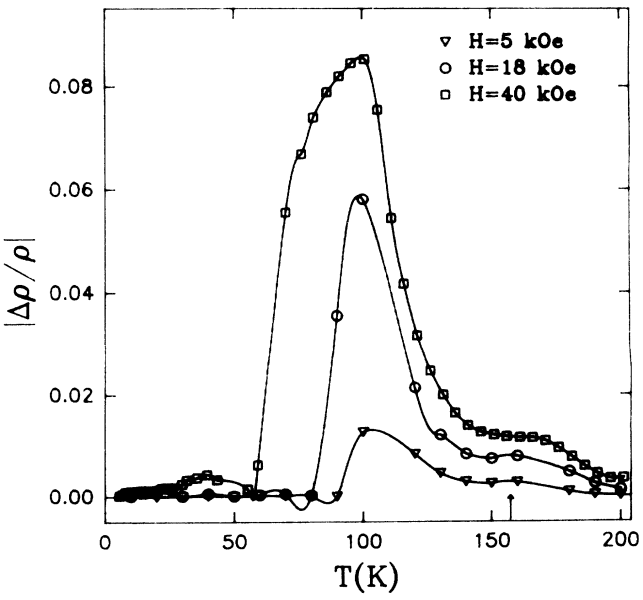

FIG. 4. Temperature (T) dependence of the magnetoresistance ($\Delta\rho/\rho$) in $Ce(Fe_{0.92}Al_{0.08})_2$ at different fields.

It has been suggested in an earlier theoretical work[17] that the Al atoms around Fe-Fe bonds can induce AF coupling between the Fe moments. The effective interaction in such a case is of Ruderman-Kittel-Kasuya-Yosida (RKKY) type: $\cos(k_F R_{ij}+\phi)/R_{ij}^3$, where R_{ij} is the distance between the Fe impurities and ϕ is the phase shift associated with the Al impurity. In case of strongly perturbative impurities like Al, ϕ will be large enough to bring about a change in the sign of the interaction leading to an antiferromagnetic coupling between Fe moments.

The temperature dependence of the ordered magnetic moments on Fe sites shows that the AF component develops at a temperature below the ferromagnetic ordering temperature.[5] This AF component begins to suppress the ferromagnetic response to the external field which leads to a drop in susceptibility.[3] In the $Ce(Fe_{0.92}Al_{0.08})_2$ compound, it is found that the FM component develops at around 170 K and the AF component appears below 150 K.[5] Below 100 K, the FM moment drops down significantly while the AF moment is stable down to the lowest temperature. The magnetoresistance data reported here conform to this picture where we find that the AF phase undergoes a field-induced metamagnetic transition below 100 K. Above 100 K, the FM moment becomes significant while the AF moment drops. In the temperature range between 80 and 150 K there is a coexistence of FM and AF moments which presumably leads to a canted state. In this temperature range, the spin-disorder scattering responsible for the magnetoresistance behavior decreases with increasing temperature, as the FM ordered state is approached. This is reflected as a rapid drop in $|\Delta\rho/\rho|$ in this temperature range. The peak in $|\Delta\rho/\rho|$ is independent of the applied field though the temperature at which negative MR develops is different for various applied fields (Figure 4). This is due to the fact that the field for the metamagnetic transition varies with temperature as shown in Fig. 3. Above 150 K, there is no coexistence phase and one has only the FM phase.[5] A small hump in $|\Delta\rho/\rho|$ is observed at the FM ordering temperature (170 K) determined from neutron-scattering measurements.[5] This apparently occurs as a result of scattering due to spin-wave excitations which increases with temperature.

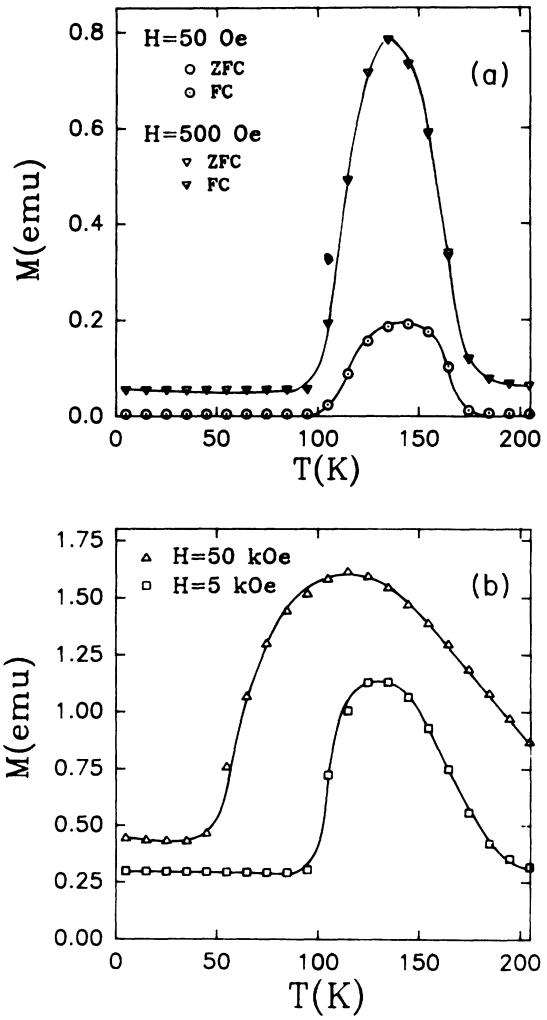

FIG. 5. Magnetization (M) as a function of temperature (T) in $Ce(Fe_{0.92}Al_{0.08})_2$ in magnetic fields of (a) 50 Oe and 500 Oe and (b) 5 kOe and 50 kOe. Measurements at 50 Oe and 500 Oe were carried out both in zero-field-cooled (ZFC) as well as field-cooled (FC) conditions. ZFC and FC data coincide; hence the data points under the two conditions are indistinguishable.

C. Temperature dependence of the magnetization

The temperature dependence of the magnetization on the same sample in fields of 50 Oe to 50 kOe is shown in Figs. 5(a) and 5(b). At 50 Oe and 500 Oe, the data were taken both in field- and zero-field-cooled conditions. The data do not exhibit any deviation between field- and zero-field-cooled conditions and hence rules out the spin-glass behavior in the compound as reported earlier.[4] In low dc fields (50 Oe), the magnetization shows a peak around the same temperature as observed in ac susceptibility.[3] This peak moves to lower temperatures with increasing fields; e.g., it is at 150 K in a field of 50 Oe while it is at 115 K in 50 kOe. However, it is interesting to note here that the peak in $|\Delta\rho/\rho|$ is independent of external field.

IV. CONCLUSIONS

The present study of magnetoresistance and magnetization shows that the $Ce(Fe_{0.92}Al_{0.08})_2$ compound undergoes an inhomogeneous ferromagnetic transition at around 170 K and has an antiferromagnetic state below 100 K. These macroscopic measurements along with the existing neutron diffraction results[5] strongly suggest that the ideas of a spin glass and reentrant spin glass[4] are not relevant for the present system. Such a view gets further support from a recent nonlinear susceptibility study on some $Ce(Fe,Al)_2$ compounds.[8] Our results also indicate that among the macroscopic measurements, the magnetoresistance study provides a better identification of various magnetic transitions occurring in the present type of systems.

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