Magnetic instability in $CeFe_2$: Effects of Re and Ir substitutions

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We present here the resistivity and magnetization study of Ir and Re substituted *C*-15 Laves-phase compound $CeFe_2$. Re and Ir substitution triggers the incipient ferromagnetic instability in $CeFe_2$ resulting in a transition from ferromagnetic to antiferromagnetic state. Both resistivity and magnetization show clear indications of both paramagnetic-to-ferromagnetic and ferromagnetic-to-antiferromagnetic transitions. The antiferromagnetic phase is restored to ferromagnetic phase on application of sufficiently strong magnetic field. [S0163-1829(97)06137-7]

Interesting physical properties of the C-15 Laves-phase compound $CeFe_2$ have been highlighted in recent years through both experimental[1,2] and theoretical[3] studies. It is now recognized that the low temperature ($T<50$ K) magnetic state of this ferromagnetic ($T_c \approx 235$ K) compound is on the verge of a magnetic instability.[4,5] The hint of such a behavior already existed from detailed experimental studies[6–12] on $Ce(Fe_{1-x}T_x)_2$; $T=$Al, Co, and Ru pseudobinary systems. It was observed that a very small (1–3 %) substitution of Al, Co, or Ru readily gives rise to a low temperature spin-canted state, which becomes distinctly antiferromagnetic by about 5% substitution. The ferromagnetic-to-antiferromagnetic transition can be quite sharp or gradual depending on the dopant elements.[7,9] The latter kind of behavior some time gave rise to the confusion that the lower temperature magnetic state might be a spin glass or reentrant spin glass.[13] This question has now been settled decisively both through bulk properties[14–17] and neutron measurements.[12]

From comparison of the results obtained from various bulk property (e.g., dc magnetization, ac susceptibility, resistivity, magnetoresistance, specific heat, etc.) measurements, it turns out that among the macroscopic probes magnetoresistance study provides the clearest indications of the multiple phase transition observed in $Ce(Fe,Al)_2$ pseudobinaries.[15,16] It was earlier observed that the low temperature antiferromagnetic state is quite sensitive to the applied magnetic field and the ferromagnetic state could be revived on application of a moderately strong field.[6] Such a field induced ferromagnetic transition or the metamagnetic transition was clearly observed in the magnetoresistance measurements as well.[15,16] A clear picture of the ferro- to antiferromagnetic transition is also obtained in $Ce(Fe,Ru)_2$ pseudobinary systems from a very recent magnetoresistance measurement.[18] An additional interesting feature which emerged from this latter study is that a relatively large magnetoresistance is associated with the field induced spin realignment process. These new results have motivated us to search for new $CeFe_2$ pseudobinary systems showing ferro- to antiferromagnetic transition, which might have even larger magnetoresistance. Also it is now clearly known that the observed multiple magnetic transitions in $CeFe_2$ based pseudobinaries are not due to disorder induced effects of the Fe sublattice; substitution with Ni, Pd, Pt, Rh, and Mn produce simple dilution effects, namely T_c decreased monotonically without any hint of a second magnetic transition.[9,10] So discovery of pseudobinary systems showing multiple magnetic transition will also be useful in building a phenomenological model to explain the anomalous magnetic properties of $CeFe_2$. Existing theoretical study[19] and some preliminary experimental results[20] indicate that substitutions with Re, Os, and Ir might induce the double magnetic transitions in $CeFe_2$. In the following sections we shall present results of our detailed resistivity and dc magnetization measurements on $Ce(Fe_{1-x}T_x)_2$ systems where $T=$Re and Ir and $0.03 \leqslant x \leqslant 0.08$. Our results indicate that these elements trigger readily the magnetic instability in $CeFe_2$ and we shall compare our findings with similar effects observed in other $CeFe_2$ based pseudobinaries.

The pseudobinary compounds used in the present study were prepared by argon arc melting from metals of nominal 99.99% purity and subjected to the following annealing procedure: 600 °C for 2 days followed by 700°C for 5 days, 800°C for five days and 850°C for one day. The samples were characterized by x-ray diffraction and metallographic studies. While x-ray studies revealed only lines identifiable with C-15 Laves-phase structure, a very small amount of second phase ($\leqslant$ 2%) could be observed in the metallography. The magnetization measurements were performed using a commercial SQUID magnetometer (Quantum Design MPMS5). The samples used are of typical dimensions of 3 mm$\times$1 mm$\times$1 mm and they were mounted in the sample holder with the long axis parallel to the magnetic field. A scan length of 4 cm was used and the measurements were averaged over three such scans each containing 32 data points. The resistivities of the rod shaped sample (10 mm $\times$1 mm$\times$1 mm) were measured between 77 and 300 K.

In Figs. 1 and 2 we present magnetization (M) vs temperature (T) plots of $Ce(Fe_{1-x}Re_x)_2$ and $Ce(Fe_{1-x}Ir_x)_2$ with $x=0.02$, 0.05, 0.07, and 0.08 for Re and $x=0.03$, 0.05, and 0.08 for Ir measured in an applied field of 100 Oe. It is clear from Figs. 1 and 2 that the lower temperature magnetic transition sets in quite readily with both the Re and Ir substitution; signature of the second transition is clearly visible with $x=0.03$ in the case of Ir substituted samples and becomes quite sharp by the time one reaches $x=0.05$. Such a behavior is quite similar to what has been observed in

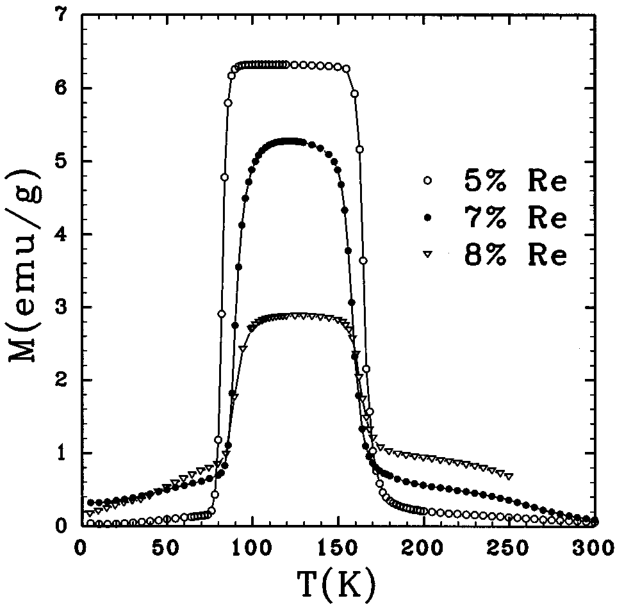

FIG. 1. Magnetization as a function of temperature in $Ce(Fe_{1-x}Re_x)_2$ for $x=0.02$, 0.05, 0.07, and 0.08. The paramagnetic to ferromagnetic transition temperature falls as x rises above 0.02, while the ferromagnetic to antiferromagnetic transition temperature (not seen in $x=0.02$) rises as x rises. The measurements were carried out in an applied field of 100 Oe.

$Ce(Fe,Ru)_2$ systems. In the case of Ru and Al substituted $CeFe_2$ the minimum concentration where the lower transition is observed is about 2% in Ru[10] and 2% in Al[9] samples. The minimum concentration of 2% observed in Ir and Re substituted compounds also compare well with this. The sharpness of the lower temperature in Ir and Re samples are comparable to that of Ru substituted and not a gradual one as observed in the Al sample. The concentration (≈8%) of dopants for which the two transitions are merged resulting in a

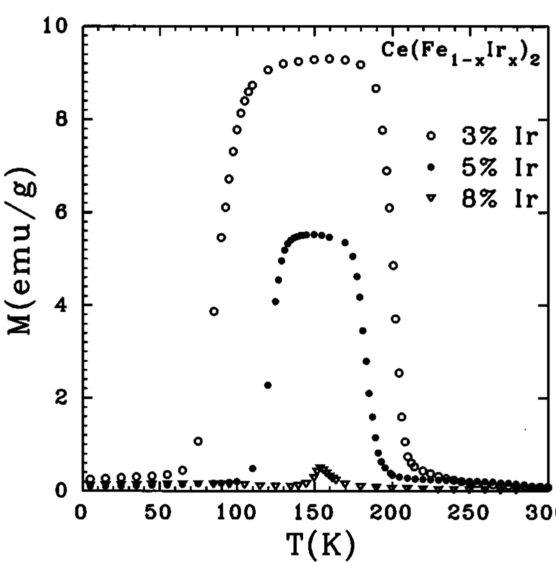

FIG. 2. Magnetization as a function of temperature in $Ce(Fe_{1-x}Ir_x)_2$ for $x=0.03$, 0.05, and 0.08. The measurements were carried out in an applied field of 100 Oe.

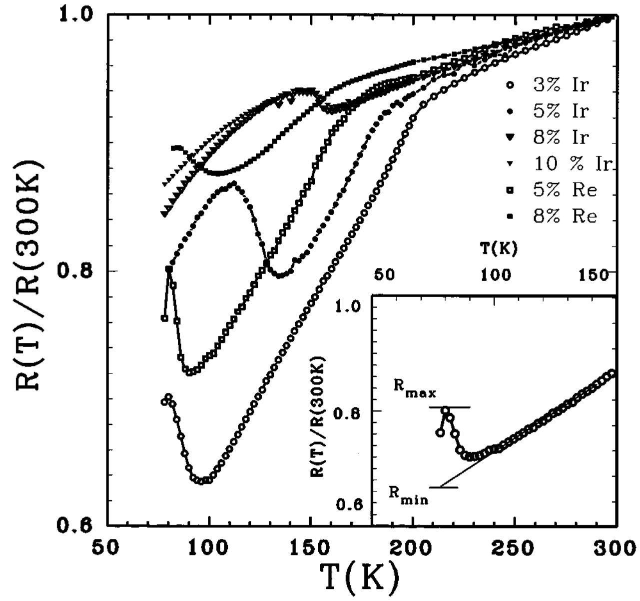

FIG. 3. Normalized resistance $R(T)/R(300)$ as a function of temperature in $Ce(Fe_{1-x}Re_x)_2$ for $x=0.05$ and 0.08 and in $Ce(Fe_{1-x}Ir_x)_2$ for $x=0.03$, 0.05, 0.08, and 0.1. The change of slope near 200 K signals the para- to ferromagnetic transition. The sharp rise in the resisitance near 90 K in $x=0.05$ and near 100 K in $x=0.08$ samples mark the ferro- to antiferromagnetic transition. The inset shows the R_{min} and R_{max} for estimating the magnetoresistance.

peaklike structure in magnetization is also comparable to that of Ru substituted samples whereas in Al samples this takes place at a much less concentration of 5%.

In Fig. 3 we present resistivity (ρ) vs T plots of $Ce(Fe_{1-x}Re_x)_2$ and $Ce(Fe_{1-x}Ir_x)_2$ with $x=0.05$ and 0.08 for Re and $x=0.03$, 0.05, 0.08, and 0.10 for Ir. The $CeFe_2$ based pseudobinaries in general are quite brittle in nature and the samples used in the present study are no exception either; they contain a number of microcracks. This causes inconsistency in the measured absolute values of the resistivity and hence we decided to plot in figures the normalized value of the resistivity $[R(T)/R(300\text{ K})]$. The para- to ferromagnetic transitions in various alloys is indicated by the sharp knee in the ρ vs T plot which is indicative of the decrease in spin disorder scattering due to the onset of ferromagnetic ordering. T_c's obtained from resistivity measurements agree well with those obtained from magnetization measurements. The lower temperature magnetic transition is identified with the sharp structure in the form of a local minimum which has been associated with superzone boundary effects in other $CeFe_2$ based pseudobinaries.[9,10] Such a behavior can be rationalized in terms of the models used to discuss the resistivity behavior of rare-earth metals.[21,22] As was discussed earlier in the case of $Ce(Fe,Al)_2$ and $Ce(Fe,Ru)_2$ pseudobinary systems[9,10] while the same Brillouin-zone structure is appropriate for the conduction electrons in the paramagnetic and ferromagnetic conditions, the superzone boundaries that appear on antiferromagnetic ordering cause a remapping of the Fermi surface which in turn reduce effective freedom of the conduction electrons and increase the resistivity. If the magnetic order is already well established by the time the

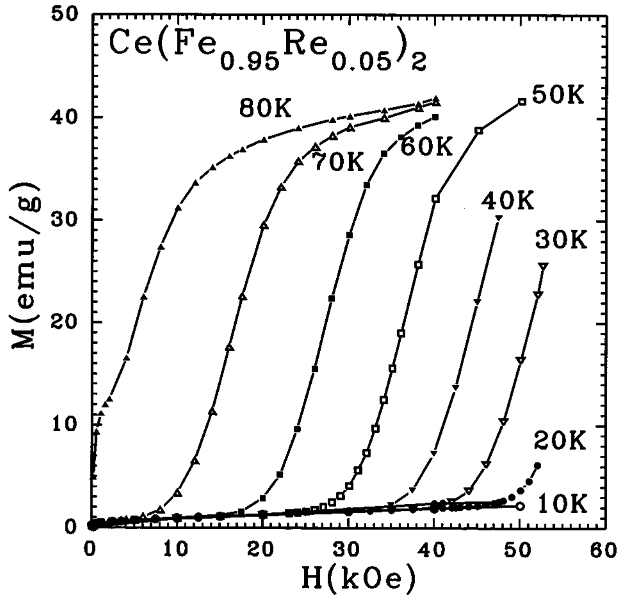

FIG. 4. Magnetization vs applied field of $Ce(Fe_{0.95}Re_{0.05})_2$. The sharp increase in magnetization indicates the onset of metamagnetic transition.

transition is reached, the increase in the resistivity is fairly sharp.[10] In the present case the 8% Re and 5% Ir doped samples display a relatively gradual rise in $\rho(T)$ at T_N. Apparently T_N of these samples lie in a temperature region where the sublattice magnetization is still increasing with the decrease in temperature.

In Figs. 4 and 5, we present results of our magnetization measurements on $Ce(Fe_{1-x}Re_x)_2$ and $Ce(Fe_{1-x}Ir_x)_2$ samples. Below T_N we find that the magnetization at low fields rises initially quite rapidly before tending to saturate with a relatively small slope. Then at a higher field (which depends on temperature) magnetization rises very rapidly indicating the onset of a field induced ferromagnetic or metamagnetic transition. The initial increase in the low field magnetization has also been observed in $Ce(Fe,Al)_2$ and

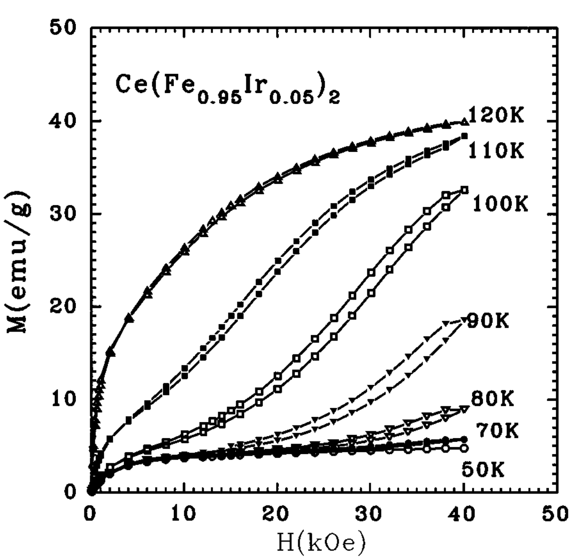

FIG. 5. Magnetization vs applied field of $Ce(Fe_{0.95}Ir_{0.05})_2$.

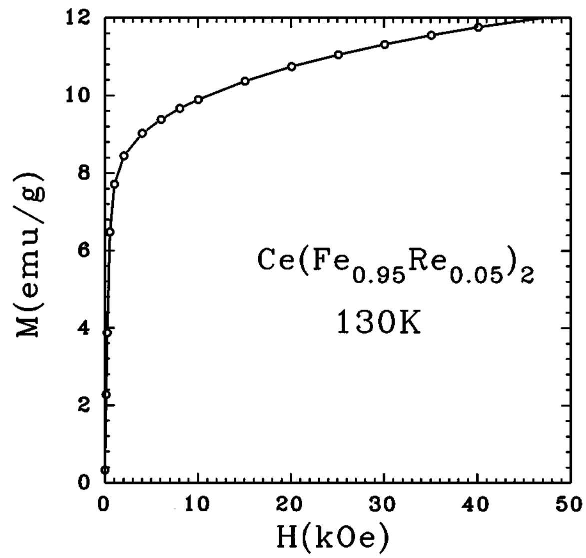

FIG. 6. Magnetization vs applied field of $Ce(Fe_{0.95}Re_{0.05})_2$ at 130 K (in the ferromagnetic regime).

$Ce(Fe,Ru)_2$ (Refs. 9 and 10) pseudobinaries. Kunkel *et al.*[18] argued that such a behavior may originate from the departure from collinearity among the Fe spins induced by the perturbing potential at the dopant sites. A deviation from the linear rise in the magnetization in the antiferromagnetic phase may also occur due to a small amount of ferromagnetic impurity phase which can easily evade detection in the standard x-ray diffraction measurements.

The metamagnetic transition in the *M*-*H* plots is accompanied by marked hysteresis which increases in strength as the temperature is lowered. We believe that this kind of hysteresis reflects the first-order nature of the metamagnetic transition. Had this hysteresis been due to the domain wall pinning in the ferromagnetic state, then a similar characteristic would have existed in the ferromagnetic state observed in the temperature regime $T_N<T<T_c$. Figure 6 shows the *M* vs *H* plot in such a temperature regime which, on the contrary, indicates the ferromagnetism is quite soft in character with a coercivity field of about 10 Oe (at $T=120$ K). This indicates that the impurity potential which can pin the domain walls is pretty weak in the temperature regime ($T_N<T<T_c$) and there is no reason to believe that the strength will change abruptly with lowering down the temperature by 20 K only. Another possible source for the observed hysteresis is magnetocrystalline anisotropy, and there is no indication that it is playing an important role here. Also the indication of the difficulty of domain movements is usually obtained in the form of strong thermomagnetic irreversibility obtained in the form of the difference between zero field cooled (ZFC) and field cooled (FC) magnetization measurements. In our temperature scans of magnetization we have not observed much difference between the ZFC and FC magnetizations.

It is already known that this ferromagnetic to antiferromagnetic transition is quite sensitive to the applied magnetic field and the sharp rise in resistivity T_N can be suppressed with an applied field of H_m or in other words T_N can get pushed down in temperature. This, in principle, gives rise to substantial magnetoresistance, which has now actually been observed by Kunkel *et al.*[18] in the Ce(Fe, Ru)$_2$ pseudobinary system. Kunkel *et al.* reported a magnetoresistance value of 22% at $T=80$ K in an applied field of 7.2 T. Since it is the suppression of the rise in the $\rho(T)$ at T_N in an applied field which gives rise to large magnetoresistance, a simple comparison of this rise $(R_{\mathrm{max}}-R_{\mathrm{min}})/R_{\mathrm{min}}$ in Ce(Fe,Ru)$_2$ pseudobinaries[18] with those obtained in our present study on Re and Ir based $CeFe_2$ pseudobinaries suggests that the magnetoresistance obtained in Ce(Fe,Re)$_2$ can be of the order of 25%. [In the estimation of the magnetoresistance, we have taken into account the change in the resistivity $\Delta\rho$ which comprises the rise in the resistivity in the narrow temperature range $\Delta\approx10$ K and extrapolated value of the decrease in resistivity over this temperature range in the absence of the antiferromagnetic transition (inset in Fig. 3).]

In summary we have found that as in the case of Ru and Al doping, substitution of $5d$ elements Ir and Re readily trigger the incipient magnetic instability of $CeFe_2$. In conjunction with the existing theoretical study, the present work again suggests the possible correlation between the interesting electronic structure of this compound and the magnetic properties. The ferro- to antiferromagnetic transition in these $5d$-element-doped $CeFe_2$ pseudobinaries is also quite sensitive to the applied magnetic field. And on comparison with the electrical and magnetic properties of the similar Ru-doped $CeFe_2$ pseudobinaries, we predict that a relatively large magnetoresistance will be observed especially in Ce(Fe,Re)$_2$ systems.

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