The nature of 4f electron magnetism in the diluted ferromagnetic Kondo lattice, CeIr$_2$B$_2$

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Abstract

We report on the physical properties of the series Ce$_{1-x}$La$_x$Ir$_2$B$_2$ (x = 0–0.9), obtained by means of magnetization, heat capacity and electrical resistivity measurements as a function of temperature (down to 0.7 K for the latter two measurements). The Curie temperature of CeIr$_2$B$_2$ (∼5 K) is lowered due to La substitution, as expected. However, no quantum critical point or ‘non-Fermi liquid’ behavior was observed even in the dilute limit of x = 0.9. Interestingly, ferromagnetic ordering persists even for Ce$_{0.1}$La$_{0.9}$Ir$_2$B$_2$, below 0.8 K. Among the Ce systems, CeIr$_2$B$_2$ is one of the compounds in which direct 4f–4f interaction does not appear to play any role in the magnetism, which is controlled by the indirect exchange interaction alone. In this compound, the Kondo effect persists in the ferromagnetic ordered state, as inferred from the entropy data.

(Some figures may appear in colour only in the online journal)

1. Introduction

The Ce-based intermetallics are taking center stage in research aimed at probing the competition between the intersite magnetic interaction and the on-site Kondo interaction [1, 2]. Several exotic properties have been discovered, attributable to this competition. The strength of the hybridization can be tailored via various parameters like the composition (x), external pressure (P) and magnetic field (H) [3–5]. It is often recognized that, besides these factors, direct 4f–4f interaction also plays a crucial role in deciding the properties in the case of Ce systems and it coexists with the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction [6, 7].

In this paper, we focus on the compound CeIr$_2$B$_2$, which exhibits ferromagnetic (FM) ordering around (T$_C$ =) 6 K [8]. Though this behavior was reported more than a decade ago, further extensive investigation is lacking in the literature. Hence, it would be interesting to probe how the low temperature properties of Ce evolve with the suppression of $T_C$ due to a change in chemical environment, through the dilution of the Ce sublattice. For this purpose, we choose to gradually replace Ce with La. It is observed that, while the $T_C$ can be gradually lowered in temperature with the dilution of the Ce sublattice, there is evidence for the persistence of magnetic ordering even in Ce$_{0.1}$La$_{0.9}$Ir$_2$B$_2$, implying that indirect RKKY interaction alone, rather than intersite interaction, determines the magnetism of this series.

2. Experimental details

The compounds, Ce$_{1-x}$La$_x$Ir$_2$B$_2$ (x = 0.0, 0.3, 0.5, 0.7 and 0.9), were prepared by arc melting stoichiometric amounts of respective high purity elements in an atmosphere of argon. X-ray diffraction studies established that all the samples are single phase (figure 1), revealing that all the compounds crystallize in a CaRh$_2$B$_2$ type orthorhombic structure. A comparison of x-ray diffraction patterns of the parent and La substituted compounds (inset of figure 1(a)) reveals a gradual shift of diffraction lines with La substitution, thereby establishing that La does indeed go to the Ce site without precipitating any other phase, within the detection limit (<2%) of this technique.

The dc magnetization ($M$) measurements (in the range 1.8–300 K) for all specimens were carried out with the help of a commercial superconducting quantum interference device.
the effect of external pressure. In order to make a comparison the magnetic transition. The $T$ for various values of the external pressure in the vicinity of dependences of the magnetization measured in a field of 5 kOe.

Following increasing unit-cell volume due to La substitution, right side of the peak in the Doniach magnetic phase diagram, a further suppression of $T$. Also, if this compound lies on the $(T = 0.7–300 \text{ K})$, were performed with the same PPMS. The electrical resistivity, a conducting silver paint was used for making electrical contacts of the leads with the samples.

3. Results and discussion

3.1. Magnetization

Figure 2(a) shows the temperature dependence of the magnetization of Ce$_{1-x}$La$_x$Ir$_2$B$_2$ under the zero-field-cooled condition, measured in a field of 5 kOe. A sudden change in the curvature of the plot reveals that the $T_c$ gets gradually shifted to a lower temperature with increase in $x$, falling below 1.8 K for $x = 0.9$. This observation is more apparent from the d$(M/H)/dT$ versus $T$ plots shown in figure 2(b). The values of $T_c$ obtained from the temperature at which there is a minimum in this plot for $x = 0, 0.3$ and $0.5$ are $\sim 5, 3.7$ and 2.5 K respectively. For the extreme compositions it is shifted below 1.8 K. The suppression of $T_c$ in this series as observed experimentally is in accordance with that expected on the basis of indirect exchange interaction. It must be emphasized that if there was a direct 4f–4f interaction, it would have led to a further suppression of $T_c$. Also, if this compound lies on the right side of the peak in the Doniach magnetic phase diagram, following increasing unit-cell volume due to La substitution, $T_c$ normalized to the Ce concentration should have exhibited an increase. Figure 2(c) shows the effect of the temperature dependences of the magnetization measured in a field of 5 kOe for various values of the external pressure in the vicinity of the magnetic transition. The $T_c$ is weakly suppressed under the effect of external pressure. In order to make a comparison with the La substituted compounds, it is desirable to know the bulk modulus of the Ce compounds, which is around 600–1000 kbar [9, 10]. From the knowledge of the unit-cell volume, it is estimated that the negative pressure exerted by the $x = 0.7$ compound corresponds to an external pressure of 7 kbar. But it is observed that $T_c$ shifts from $\sim -5$ to $3.9 \text{ K}$ under a pressure of 6 kbar. If the same trend continued with negative chemical pressure, $T_c$ (normalized to the Ce concentration) would have gone up upon La substitution, in contrast to what was observed. This implies that the compound lies at the peak of the Doniach phase diagram [11].

The susceptibility curves of this series in the range 150–300 K were fitted with the modified Curie–Weiss law. The effective moment obtained from the fitting corresponds to that of trivalent Ce for $x = 0.0$ and it decreases linearly with increasing $x$. The value of the temperature independent component ($\chi_0$) for all the compositions is $\sim 10^{-4} \text{ emu mol}^{-1}$, while the paramagnetic Curie temperature ($\theta_p$) is around $\sim -12 \text{ K}$, remaining essentially unchanged across the series, and the negative sign of $\theta_p$ for a ferromagnetic Ce alloy implies that the dominant contribution to $\theta_p$ arises from the single-ion Kondo temperature. From figure 2(d), it is inferred that the nature of the $M(H)$ curve at 1.8 K curves undergoes a change with increase of the dopant concentration. The sharp rise and magnetic hysteresis observed in low fields for $x = 0.0$ is gradually suppressed with increase of $x$ and a smooth curvature of $M(H)$ curve is observed for $x = 0.7$ for the entire field range. For $x = 0.7$ and 0.9, the ordering temperature lying below 1.8 K is responsible for this observed difference in the $M(H)$ isotherm. A magnetic moment of $0.85 \mu_B/\text{f.u.}$ at 50 kOe is observed for $x = 0.0$, which decreases linearly with increasing doping (inset of figure 2(e)), as expected for the single-ionic nature of the observed magnetic moment. The observed reduced value of the magnetic moment (compared to the free Ce$^{3+}$ moment of 2.54 $\mu_B/\text{f.u.}$) is attributed to the crystal field effect. Also, $T_c$ shifting towards zero does not result in NFL features at low temperatures (e.g. $\chi \sim \log T$) for instance, for the extreme La composition (figure 2(f)), as observed often for dilute Ce or U alloys in the literature [13].

3.2. Heat capacity

The heat capacity behavior of this series as a function of temperature is shown in figure 3(a). For $x = 0.0$ an upturn is observed below 6 K in the $C(T)$ plot, consistent with magnetic long range ordering around 5 K. This peak shifts to a higher temperature in a field of 50 kOe, implying that the magnetic ordering in this compound is FM (figure 3(b)). With the increase in doping, the features shift to lower temperatures and move below 1.8 K for $x = 0.7$ in zero field. Hence, for this composition, the $C$ measurement was extended to a further lower temperature using an He-3 insert. A clear peak around 1.4 K is observed in the $C$ versus $T$ plot of the compound (figure 3(c)). For this concentration, a peak is observed in 50 kOe at a higher temperature (4 K), implying that the FM ordering is persisting. For $x = 0.9$, a tendency for the formation of a peak is observed below 0.8 K in zero field for $C$ (figure 3(d)) without any peak at higher temperatures. When...
Figure 2. (a) Temperature ($T$) dependence (1.8–300 K) of the magnetization ($M$) divided by the magnetic field ($H$) for Ce$_{1-x}$La$_x$Ir$_2$B$_2$ for the zero-field-cooled condition. (b) Temperature response of $d(M/H)/dT$ for all of the compounds. (c) Temperature response of $d(M/H)/dT$ for all external pressures. Inset: $M/H$ versus $T$ plot under the influence of external pressure. (d) Isothermal magnetization behavior of Ce$_{1-x}$La$_x$Ir$_2$B$_2$ at 1.8 K. (e) Magnetization value at 1.8 K at 50 kOe field, plotted as a function of the La concentration. A straight line is drawn through data points. (f) $M/H$ curve plotted as a function of log $T$ for $x = 0.9$.

a field of 50 kOe is applied, $C(T)$ develops a peak around 1.6 K, implying that even for this extreme composition of this series, in this study, FM ordering persists. Also, we would like to mention that the variation of $C/T$ is not proportional to log $T$, implying an absence of NFL behavior, as inferred from magnetization measurements.

For this series, the magnetic part of $C$ ($C_{4f}$) was obtained by subtracting the lattice part, with La$_2$Ir$_2$B$_2$ as the reference for the lattice contribution. For this analysis, the difference in Debye temperatures of the series and the reference compound is taken into account using a procedure given in [12]. Also the magnetic entropy ($S_{4f}$) was estimated from the 4f contribution to $C$. For the parent compound (figure 3(e)), at $T_C$, $S_{4f} = 3.1$ J mol$^{-1}$ K$^{-1}$ Ce is $\sim$53% of that expected for the crystal field splitting of the doublet ground state. This observed lower value of the entropy implies a partial screening of the Ce magnetic moment by conduction electron spins. Hence this result implies a coexistence of the Kondo effect with FM ordering. A similar phenomenon has also been observed for the Ce series, namely Ce$_y$La$_{1-y}$Ge$_2$ [14]. Under the assumption that the reduction in entropy value is Kondo derived, an estimate of the single-ion Kondo temperature $T_K$ can be acquired from

$$S_{4f}(T_C) = S_K(T_C/T_K)$$

where $S_{4f}$ and $S_K$ are the magnetic and Kondo entropy respectively at $T_C$ [15]. According to the spin 1/2 Kondo model, $T_K$ for the parent compound was found to be around 8 K. The linear coefficient of the specific heat ($\gamma$) was obtained from a $C_{4f}/T$ versus $T^2$ plot (in the linear $T$ region above $T_C$, which is different for different compositions). The value turns out to be $\sim$150 mJ mol$^{-1}$ K$^{-2}$ for the parent compound, implying that CeIr$_2$B$_2$ is a moderately heavy fermion compound. The value of $\gamma$ decreases and it is $\sim$24 mJ mol$^{-1}$ K$^{-2}$ for $x = 0.9$, as expected for magnetic dilution.
3.3. Electrical resistivity

Figure 4 shows $\rho$ plotted as a function to $T$. We have plotted the normalized resistivity (at 300 K), as the absolute values are not very reliable due to microcracks in the specimens. For the two extreme compositions, $x=0.7$ and 0.9, $\rho$ measurements were performed up to 0.7 K. For the parent compound, a significant drop of $\rho$ around 5 K is observed and this temperature coincides with the minimum in $d\rho/dT$. This temperature is clearly the magnetic ordering temperature. For all the compositions, the temperature of the drop nearly coincides with the ordering temperature of the respective members. Also, no non-Fermi liquid signature is observed even in the dilute limit, as such a behavior can be expected for an alloy at the quantum critical point, and not for one near the peak in the Doniach magnetic phase diagram [11].

4. Conclusion

The influence of dilution of the Ce sublattice on the magnetic behavior of CeIr$_2$B$_2$ has been investigated. The $T_C$ around 5 K observed for CeIr$_2$B$_2$ is reduced gradually, with a prevalence of finite $T_C$ (below 0.8 K) even for $x=0.9$. If direct f–f interaction played a role, $T_C$ would have decreased more sharply with $x$. Thus this is an interesting Ce system in which magnetization is controlled by indirect exchange interaction alone. And finally, the results establish that CeIr$_2$B$_2$ is a ferromagnetic Kondo lattice.

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