La$_{1-x}$Ce$_x$B$_6$ (x≤0.1): Spin-polaron regime in dilute magnetic system

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Abstract. Precision measurements of charge transport characteristics (resistivity, magnetoresistance, Hall and Seebeck coefficients) and magnetic susceptibility have been carried out on high-quality single-crystals of the so-called dilute Kondo system La$_{1-x}$Ce$_x$B$_6$ (x≤0.1) in a wide temperature range 1.8-300 K. It is shown that the low temperature magnetic contribution in resistivity obeys the power law $\Delta \rho \sim T^{-\alpha}$, which corresponds to the regime of weak localization of charge carriers with the critical exponent values $\alpha=0.47-0.49$ for cerium concentrations x≤0.1. Simultaneously an asymptotic behavior of Seebeck coefficient $S \sim -\ln T$ is found together with nearly constant and negative Hall coefficient $R_H(T)$ in these dilute magnetic systems. The results of comprehensive analysis contradict to the predictions of Kondo-impurity approach for this archetypal strongly correlated electron system in dilute impurity limit. An alternative interpretation of the properties of La$_{1-x}$Ce$_x$B$_6$ is developed and based on spin-polaron approach, Pauli paramagnetism and the density of states renormalization effects at low temperatures.

1. Introduction.
Recent discovery of quantum critical behavior in archetypal heavy fermion solid solutions La$_{1-x}$Ce$_x$B$_6$ in the regime x≤0.6 [1] has brought renewed interest to study both the intriguing magnetic phase diagram and unusual properties of these strongly correlated electron systems. It was shown in [1] that special attention should be paid to the low concentration limit, particularly, x≤0.1 range, where well-known Kondo-impurity approach was generally applied.

On the other hand it was demonstrated very recently in the studies of charge transport and magnetic characteristics of the so-called dense Kondo system CeB$_6$ that, instead of the Kondo-lattice interpretation, spin-polaron approach may be considered as alternative and the most appropriate
description of the properties of cerium hexaboride [2]. Hence, to link together the dense (CeB₆) and dilute (x ≤ 0.1) limits it is promising to investigate and analyze charge transport anomalies in La₁₋ₓCexB₆ in the framework of the different scenarios.

2. Experimental results and discussion.
To shed more light on the nature of the crossover between the dilute and dense limits in La₁₋ₓCexB₆, the precision measurements of charge transport (resistivity, magnetoresistance, Hall and Seebeck coefficients) and magnetic characteristics have been carried out on the high-quality single crystals of La₁₋ₓCexB₆ (x ≤ 0.1) in a wide temperature range 1.8-300 K in magnetic field up to 70 kOe.

Figure 1 shows the resistivity temperature dependencies both for dilute magnetic alloys La₁₋ₓCexB₆ with x=0.03 and 0.1 and for nonmagnetic reference compound LaB₆ at H=0 and 70 kOe. Large positive magnetoresistance (MR) in LaB₆ changes to the pronounced negative MR effect for La₁₋ₓCexB₆ solid solutions at low temperatures (figure 1). The magnetic contribution to resistivity at H=0 was deduced (subtracting the dependence for non-magnetic reference hexaboride LaB₆ from the ρ(T)) for x=0.03 and 0.1 dilute compounds and presented on figure 2(a) together with the Δρ(T) data for dense magnetic system CeB₆. The data in figure 2(a) demonstrate evidently that at low temperatures the magnetic component obeys the power law Δρ~T⁻φ, which corresponds to the regime of weak localization of charge carriers (see, e.g., [3]) with the critical exponent values φ=0.47-0.49 for dilute compounds x≤0.1 and φ=0.39 for CeB₆ [2]. It should be stressed here that it is not possible to fit the Δρ(T) data in a wide enough range of temperatures by the logarithmic dependence Δρ~−lnT. Moreover, the problem of the Kondo-type fitting Δρ(T)~−lnT can be definitely recognized in the results of Samwer and Winzer [4] and Sato et al. [5] as well.

![Figure 1. The temperature dependencies of resistivity of La₁₋ₓCexB₆ (x=0, 0.03, 0.1) for H=0 kOe and 70 kOe.](image)

For CeB₆, the resistivity and Seebeck coefficient behavior was analyzed in detail in combination with Hall effect and magnetization data in [2]. It was evidently established that the onset of the power law behavior of resistivity is accompanied by logarithmic dependence of Seebeck coefficient S~−lnT (see also figure 2(b)) which is very unusual for metallic system. Moreover, it was shown in [2] that the
Figure 2. The temperature dependencies of (a) magnetic contribution to resistivity $\Delta \rho$ (see text) and (b) Seebeck coefficient $S(T)$ in La$_{1-x}$Ce$_x$B$_6$.

Figure 3. The temperature dependencies of (a) the Hall coefficient $R_H(T)$ and (b) magnetic contribution to susceptibility $\chi(T)$ in La$_{1-x}$Ce$_x$B$_6$. 
logarithmic increase of thermopower in CeB₆ at intermediate temperatures should be attributed to the dramatic density of states (DOS) renormalization (formation of spin-polaron type manybody resonance in DOS at $E \leq E_F$). At present study the similar logarithmic asymptotic $S \sim -\ln T$ was detected also for dilute solid solutions La$_{1-x}$Ce$_x$B₆ with $x \leq 0.1$ at temperatures 10-40K (figure 2(b)). Additionally it is easy to deduce from the data of figure 2(b) that the increase of concentration $x$ in La$_{1-x}$Ce$_x$B₆ resulted in (i) an enhancement of the thermopower values, (ii) a rise of the slope of $S \sim -\ln T$ dependence and (iii) an expansion of the logarithmic law temperature interval. Then, the Hall coefficient $R_H(T)$ in the dilute $x=0.03$ and 0.1 (figure 3(a)) and dense (CeB₆, see, for example, [2]) La$_{1-x}$Ce$_x$B₆ magnetic systems is negative and it is nearly constant (a variation $\Delta R_H/R_H(300K) \leq 6 \%$) in the wide temperature range 5-300 K. Moreover, the $R_H(T)$ behavior is quite similar to that one observed for nonmagnetic reference compound LaB₆ (figure 3(a)) which is contrary to the prediction of the skew-scattering and Kondo-lattice models [6-7].

It was also shown in [2] that the magnetic susceptibility temperature dependence $\chi(T) \sim T^{-\beta}$ with index $\beta \approx 0.78$ correlates with the charge carriers’ effective mass variation $m_{eff}(T) \sim T^{-\beta}$ in CeB₆ at intermediate temperatures, resulting from the DOS renormalization effects and, hence, an enhancement of the Pauli paramagnetic response in this strongly correlated electron system. To analyze the changes in $\chi(T)$ behavior between the dense and dilute regimes in the La$_{1-x}$Ce$_x$B₆ system the magnetic susceptibility measurements have been carried out in present study and the magnetic contribution $\chi(T)_{La_{1-x}Ce_xB_6} \sim \chi(T)_{LaB_6}$ was directly deduced (see figure 3(b), $\chi(T)_{LaB_6} = -5.36*10^{-5}$ emu/mole). The results of the high precision measurements allow us to establish a non-Curie-Weiss-like behavior of the magnetic contribution $\chi(T) \sim T^{-\beta}$ with $\beta<1$ even in the case of a small enough Ce impurity concentration $x=0.03$. It should be also mentioned that an unusual correlation $2\alpha = \beta$ between the exponents $\alpha$ for resistivity (figure 2(a)) and $\beta$ for magnetic susceptibility (figure 3(b)) of the La$_{1-x}$Ce$_x$B₆ was found both for the dense and dilute compounds under investigation.

Finally, it was shown that the data obtained in the resistivity (figure 1, 2(a)), thermopower (figure 2(b)), Hall coefficient (figure 3(a)) and magnetic susceptibility (figure 3(b)) studies of La$_{1-x}$Ce$_x$B₆ ($x \leq 0.1$) in combination with magnetoresistance results [8] contradict to the predictions of Kondo-impurity approach for the archetypal strongly correlated electron system in low impurity limit. An alternative interpretation of the properties of La$_{1-x}$Ce$_x$B₆ is developed and based on spin-polaron approach, Pauli paramagnetism and DOS renormalization effects at low temperatures.

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References
[1] Nakamura S, Endo M, Yamamoto H, Isshiki T, Kimura N, Aoki H, Nojima T, Otani S and Kuni S 2006 Phys. Rev. Lett. 97 237204
[2] Sluchanko N E, Bogach A V, Glushkov V V, Demishev S V, Ivanov V Yu, Ignatov M I, Kuznetsov A V, Samarín N A, Semeno A V and Shitsevalova N Yu 2007 JETP 104 120
[3] McMillan W L 1981 Phys. Rev. B 24 2739
[4] Samwer K and Winzer K Z. Physik B 1976 25 269
[5] Sato N, Sumiyama A, Kuni S, Nagano H and Kasuya T 1985 J. Phys. Soc. Jpn. 54 1923
[6] Coleman P, Anderson P W and Ramakrishnan T V 1985 Phys. Rev. Lett. 55 414
[7] Hadzic-Leroux M, Hamzic A, Fert A, Haen P, Lapierre F and Laborde O 1986 Europhys. Lett., 1, 579
[8] Sluchanko N E et al., to be published elsewhere