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Spin-hole doping in the Kondo insulator $Ce_3Bi_4Pt_3$ studied by neutron scattering

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Abstract

Inelastic neutron scattering from $(Ce_{1-x}La_x)_3Bi_4Pt_3$, with $x = 0, 0.1$ and 0.25, shows that a gap in the spin excitation spectrum decreases monotonically with increasing x and remains well-defined even for $x = 0.25$.

Although the vast majority of heavy-electron compounds are metallic at low temperatures, a few are known to have small semiconducting gaps E_g on the order of tens of Kelvins. A simple interpretation for the origin of E_g is that the bare 4f- or 5f-energy level is renormalized by many-body interactions to be degenerate with a broad conduction band with which the renormalized f-level hybridizes. Provided the electron count, including the strongly interacting f-electrons, is exactly two, the lower hybridized band will be filled and the upper band will be empty, leaving an indirect gap from zone boundary to zone center of order V^2/W, where V is the hybridization matrix element and W is the conduction bandwidth [1]. An interesting aspect of this model is that, in addition to the existence of a charge gap, there should also be a gap in the spin excitation spectrum [2, 3]. Experimental confirmation of a spin gap has come from inelastic neutron scattering on $Ce_3Bi_4Pt_3$ [4] and CeNiSn [5]. In both cases the spin and charge gaps are of comparable magnitude and of order 100 and 10 K for the two compounds, respectively.

When the f-sublattice periodicity is broken by substituting La for Ce in $(Ce_{1-x}La_x)_3Bi_4Pt_3$ a metallic-like resistivity develops, the T-linear specific heat coefficient increases from near zero to a large value characteristic of a weakly mixed-valence compound, and a peak at $T = T_{max}$ in the static susceptibility moves to lower temperatures [6]. Similar trends have also been reported in $Ce_{1-x}La_xNiSn$ [7], $Sm_{1-x}La_xB_6$ [8] and $Yb_{1-x}Lu_xB_{12}$ [9], and are consistent with the presence of Kondo-like interactions. An important question to be answered is how do these small-gap semiconductors evolve into heavy-electron metals with increasing concentrations of nonmagnetic La or Lu. To address this question we have determined the evolution of the spin gap in $(Ce_{1-x}La_x)_3Bi_4Pt_3$ by inelastic neutron scattering experiments.

Single crystals of $(Ce_{1-x}La_x)_3Bi_4Pt_3$ were grown from a Bi flux with nominal La concentrations $x = 0, 0.1, 0.25$ and 1.0. Electrical resistance and static susceptibility measurements on these crystals followed systematics reported previously [6]. Neutron scattering was performed on the HET spectrometer at the Rutherford spallation source using incident neutron energies E_0 of 30 and

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100 meV. Powdered samples, prepared by grinding the crystals, were held at 11 ± 1 K throughout the experiments to minimize the presence of thermally activated electron–hole pairs. To obtain reasonable statistics, data collected in low-angle detectors $5^\circ < \Theta < 26.5^\circ$ were added together, giving momentum transfer $Q = 1.07\,\text{Å}^{-1}$ at zero energy transfer for $E_0 = 30$ meV. Data taken with $E_0 = 100$ meV will be presented later. To determine the magnetic scattering contribution, data for $La_3Bi_4Pt_3$ were scaled by the ratio of averaged Ce and La cross-sections and subtracted. The difference was divided by the Ce concentration. All data were corrected by measuring an empty sample holder, cadmium and a vanadium standard in the sample position.

Figure 1 shows the results of these measurements, which are plotted as the magnetic scattering cross-section versus energy transfer. The $x = 0$ results are, within experimental error, identical to earlier measurements [4] that show the absence of magnetic scattering below the gap edge of ~ 13 meV, above which scattering grows rapidly to a maximum near 25 meV. With increasing La content the gap edge Δ_g moves rapidly to smaller energy transfers, decreasing approximately as $\Delta_g(x) = 12.9 - 13.6x^2$, where the numerical values have units of meV. For $x > 0$, it is possible that scattering is finite below Δ_g, but given statistical fluctuations in the data, no definitive statement can be made. It is possible that quasi-elastic scattering may develop from spin fluctuations allowed by "spin holes" created in the lower hybridized band by the replacement of nonmagnetic La for Ce. As pointed out by Riseborough [2], such intraband scattering is expected to be weak and distributed over a wide range of energies. What remains clear from these data, though, is that a gap in the spin spectral function remains well-defined even when one-fourth of the Ce atoms are replaced by La.

A more appropriate definition of the spin gap may be that energy transfer required to increase magnetic scattering to one-half its maximum value. Values of $\tilde{\Delta}_g$ so defined are noted by arrows in Fig. 1. Unlike Δ_g, $\tilde{\Delta}_g$ decreases linearly with x as $\tilde{\Delta}_g(x) = 17.7 - 26x$. For pure $Ce_3Bi_4Pt_3$ a maximum in the static susceptibility is expected at some finite temperature due to the thermal population of holes in the lower hybridized band, thereby permitting intraband excitations. The susceptibility should peak at temperatures on the order of the indirect band gap [2]. We find that the ratio $\tilde{\Delta}_g/k_BT_{max}$ is 2.5 ± 0.1, ranging from 2.56, 2.54 and 2.40 for $x = 0, 0.1$ and 0.25, respectively. (The significance of the magnitude of this ratio is not known but we note that it does correspond, perhaps coincidentally, to the value of the total angular momentum of Ce^{3+} in the absence of crystal fields, which appear [4] to be insignificant in $Ce_3Bi_4Pt_3$.) There are two important conclusions to be drawn from this observation: provided $\tilde{\Delta}_g$ is a proper definition of the magnitude of the gap in the spin excitation spectrum, then (1) as suggested theoretically [2], T_{max} does provide a measure of the gap and (2) creation of spin holes by thermal excitation or La substitution has comparable effects on the magnitude of the gap. The linear (or quadratic) decrease in the gap with increasing x appears at odds with a theoretical model [10] of Kondo holes in these small-gap semiconductors. In this model the concentration of holes does not change the magnitude of the hybridization gap; it is dominated by the intra-atomic Coulomb repulsion U, which presumably is a weak function of x. Instead, increasing La substitution builds an impurity band within the gap whose width and f-density of states are proportional to $\sqrt{x}$. Although specific heat measurements [6] on $(Ce_{1-x}La_x)_3Bi_4Pt_3$ are consistent with this prediction, the spin gap appears insensitive to this impurity band.

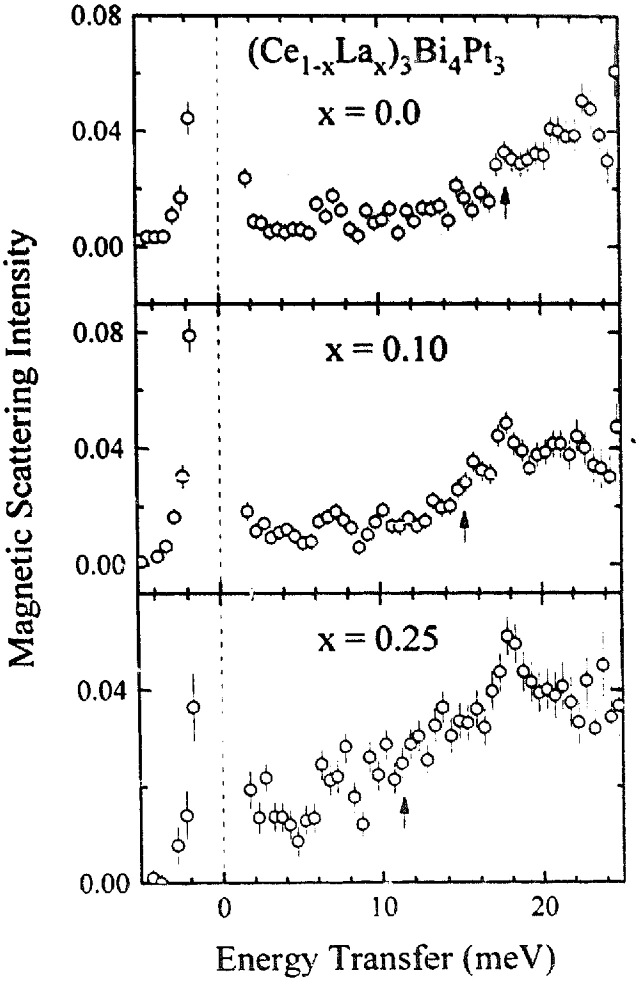

Fig. 1. Neutron magnetic scattering cross-section in arbitrary units versus energy transfer for $(Ce_{1-x}La_x)_3Bi_4Pt_3$, with $x = 0$, 0.1 and 0.25. Data have been normalized by the fractional Ce content. See text for details.

The existence of a spin gap even for $x = 0.25$ suggests that coherence effects are not significant, and that spin dynamics are determined locally. Possibly, gap suppression with increasing x is caused by negative "chemical pressure" created by expanding the lattice with La substitution, which is consistent with charge gap increasing with applied pressure [6].

In summary, we have shown for the first time that a gap in the spin function of the Kondo insulator $Ce_3Bi_4Pt_3$ decreases monotonically with increasing substitution of La for Ce and does not collapse the gap even with significant perturbation in the Ce sublattice periodicity, that Kondo-like features in resistivity and specific heat appear in the presence of a spin gap and that a measure of the relative magnitude of that gap is provided by the temperature at which the static susceptibility peaks.

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