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Spin-gap magnetic response in (Yb, Lu)B_{12}

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Abstract

To clarify the role of Yb–Yb correlations in the formation of the gap-like excitation spectrum of YbB_{12}, the spin dynamics of strongly diluted $Yb_{0.25}Lu_{0.75}B_{12}$ have been studied by inelastic neutron scattering in a wide temperature range. The data indicate that the spin gap is not suppressed by the dilution process even for large concentrations of Lu. However, the breaking-down of the Yb-sublattice periodicity leads to a strong smearing of the low-energy features and to a moderate suppression of the high-energy peak in the magnetic spectral response of YbB_{12}. The interrelation of the spin and charge gaps is discussed.

Keywords: Kondo insulator; Inelastic neutron scattering; Spin gap

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1. Introduction

The dodecaboride compound YbB_{12} belongs to the class of so-called Kondo insulators. It behaves as a metal or a semimetal with localized magnetic moments at room temperature, but becomes a nonmagnetic semiconductor with a narrow gap at the Fermi energy with decreasing temperature. Inelastic neutron scattering (INS) measurements have revealed that YbB_{12} has a magnetic excitation spectrum unlike other systems with valence instabilities [1,2]. The most intriguing feature in the magnetic response of YbB_{12} is the spin gap like behaviour below $E = 10\,\text{meV}$ at low temperature. Above this level of energy the spectrum consists of three main components: two narrow peaks at $E = 15$ and $20\,\text{meV}$, and a broad peak at $38\,\text{meV}$. Increasing temperature produces drastic changes in the spectral response: (i) the two low-energy excitations are replaced by one peak at about $23\,\text{meV}$; (ii) a substantial quasielastic peak appears, whereas the upper ($38\,\text{meV}$) inelastic peak is totally suppressed. These two contrasting forms of the magnetic response at $T \approx 15$ and $\sim 100\,\text{K}$ can, respectively, be associated with the Kondo-insulator ground state and a single-ion-type state, which is recovered with increasing temperature.

The spin gap value defined from INS is comparable to the energy of the charge gap in the electron density of states determined from transport [3], heat capacity [4], and optical conductivity [5] measurements. The relation between these two gaps is one of the central questions of the Kondo-insulator phenomenon. The theoretical models proposed for Kondo insulators are controversial and consider Yb-sublattice coherence as either essential [6] or irrelevant [7] to the gap formation. In a recent study of the effect of Lu substitution on the magnetic spectral response in YbB_{12} [8], it was concluded that Yb-sublattice coherence is the most important factor affecting the low-energy structure near the gap at low temperatures, but that the spin gap itself may have a primarily incoherent character, as it is still found to exist in the $Yb_{0.75}Lu_{0.25}B_{12}$ spectra. The high-temperature limit of the Yb magnetic spectrum is not influenced by 25% substitution and can be treated as representative of the "single-ion" spin-fluctuation regime. Surprisingly, even for the high concentration of

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non-magnetic defects (90%) the spin gap does not seem to disappear, i.e. the $Yb_{0.1}Lu_{0.9}B_{12}$ magnetic excitation spectrum does not show any observable quasi-elastic signal at low temperature. The spectrum is dominated by the broad peak centred around 38 meV, whereas the components become undetectable at 15 and 20 meV. Unfortunately, the measured intensity of the magnetic signal in $Yb_{0.1}Lu_{0.9}B_{12}$ was rather weak because of the low Yb concentration. Furthermore, the whole magnetic scattering cross-section is located in the broad peak, making the information obtained difficult to treat quantitatively. Finding clear evidence for the spin gap in a strongly diluted compound remains a major point for discussing this feature in general, and for selecting the correct approach to analysing the experimental spectra.

In this paper, we present results of a detailed INS study of the magnetic response and its temperature dependence in strongly diluted $Yb_{0.25}Lu_{0.75}{}^{11}B_{12}$. This concentration can still be assumed to represent a predominantly single-site behaviour for the Yb spin dynamics, while providing a substantial improvement in the statistical accuracy in comparison with $Yb_{0.1}Lu_{0.9}B_{12}$. We will discuss the evolution of the spin gap as a function of temperature and Lu concentration, and compare it to the corresponding evolution of the charge gap obtained from optical spectroscopy [5].

2. Experiment

Powder samples of $Yb_{0.25}Lu_{0.75}{}^{11}B_{12}$ ($m \sim 10$ g) and of the reference compound $Lu^{11}B_{12}$ ($m = 5.85$ g) were prepared at NASU Institute for Problems of Materials Science (Kiev, Ukraine) by borothermal reduction at 1700 °C under vacuum. To reduce the neutron absorption, samples were prepared using ^{11}B isotope with 99.5% enrichment. The neutron scattering experiment was performed on the time-of-flight spectrometer HET (ISIS, RAL) with two incident neutron energies $E_0 = 80$ or 50 meV, which provide resolutions (full-width at half-maximum of the elastic line in vanadium) of 3.6 and 2.0 meV, respectively. Measurements were taken at three temperatures: 10, 70, and 120 K. The experimentally defined transmission of the $Yb_{0.25}Lu_{0.75}{}^{11}B_{12}$ sample was 80% for $E_0 = 80$ meV. The $Lu^{11}B_{12}$ sample was used for estimating the phonon contribution. The magnetic part of the $Yb_{0.25}Lu_{0.75}{}^{11}B_{12}$ spectra was determined by the standard procedure using the ratio of $Lu^{11}B_{12}$ spectra for low and high scattering angles [9]. Absolute calibration was effected by normalization to a vanadium standard.

3. Results and discussion

Fig. 1 presents magnetic scattering functions for $Yb_{0.25}Lu_{0.75}B_{12}$ at the two limiting temperatures, $T = 10$ and 120 K. Data from Ref. [8] for YbB_{12} and $Yb_{0.75}Lu_{0.25}B_{12}$ are shown for comparison.

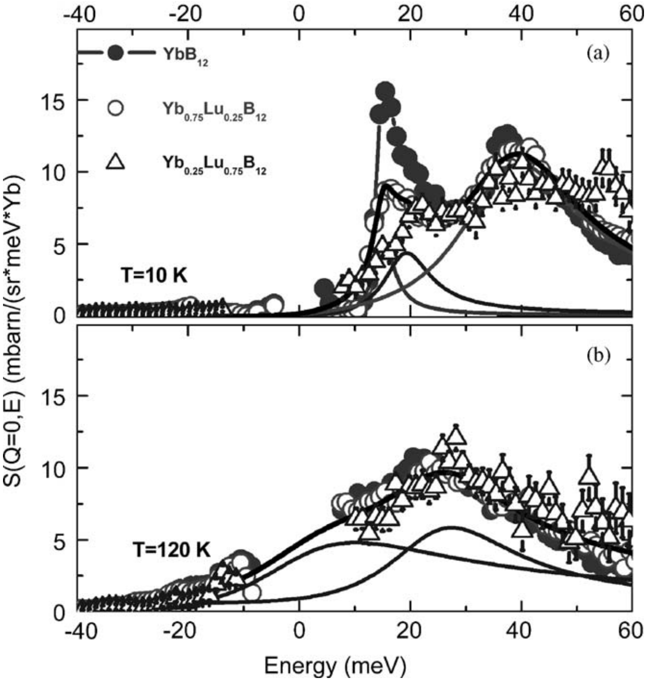

Fig. 1. Magnetic excitation spectra of $Yb_{1-x}Lu_xB_{12}$ at $T = 10$ K (a) and 120 K (b); the YbB_{12} (solid circles) and $Yb_{0.75}Lu_{0.25}B_{12}$ (open circles) spectra were measured with the incident energy $E_0 = 80$ meV [8]; the $Yb_{0.25}Lu_{0.75}B_{12}$ (triangles) spectrum was obtained as a combination of magnetic scattering spectra measured at $E_0 = 50$ and 80 meV. Lines represent a fit of the $Yb_{0.75}Lu_{0.25}B_{12}$ spectrum by the inelastic and quasi-elastic Lorentzian peaks.

The important experimental factor (Fig. 1a) is the clear evidence for the absence (or at least the strong suppression) of the magnetic signal at low temperature within the energy range of the gap regardless of Lu concentration. The three-peak structure observed in the YbB_{12} magnetic spectrum is retained in the case of $Yb_{0.75}Lu_{0.25}B_{12}$. The increase of the Lu concentration in $Yb_{1-x}Lu_xB_{12}$ from $x = 0.25$ to 0.75 results in a further broadening and a strong suppression of the near-gap low-energy peaks. This confirms that the low-energy part of the YbB_{12} inelastic magnetic spectral response at low temperature depends strongly on the concentration of non-magnetic defects.

In comparison with these near-gap components, the broad 38-meV peak appears to be much more robust with respect to the substitution (Fig. 1a). There was some indication, in the results of Ref. [8] for $x = 0.25$, of a limited change in the 38-meV peak. This peak had a shoulder at 42 meV for $x = 0$, which was not observed for $x = 0.25$. The intensity change, on the other hand, was small enough and remained within the limits of experimental accuracy. For $x = 0.75$ significant broadening is observed (Fig. 1a), and the peak shape becomes more symmetric, while its amplitude is clearly reduced.

Therefore, the breakdown of periodicity on the Yb-sublattice leads to a strong suppression of the low-energy features and a moderate change in the high-energy peak of the YbB_{12} magnetic spectral response. The important point is that no quasi-elastic scattering is observed even for