MAGNETIC SPECTRAL RESPONSE IN THE KONDO INSULATOR YbB_{12}

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Aside from the well-documented "heavy fermion superconductors", metallic Fermi liquids, or magnetically ordered materials, other strongly correlated (mixed-valence) rare-earth compounds such as SmB_6 or TmSe have been known for 30 years or so to develop an intriguing *semiconducting* ground state at low temperature. More recently, the discovery of so-called "Kondo insulators" ($Ce_3Bi_4Pt_3$, YbB_{12}) among Ce- and Yb-based compounds was a significant advance because these elements, having only one electron (or hole) on their *4f* shell, are more directly amenable to existing correlated electron theories. Experimentally, the hallmark of Kondo insulators is the gradual opening of a very narrow gap (of the order of 10 meV) in the electronic density of states at the Fermi energy when temperature decreases below ~ 100 K. This gap is thought to be a genuine effect of strong *f*-electron correlations.

YbB_{12} is an outstanding example of this type of physics: the electronic gap is observed in a number of properties (optical conductivity, photoemission spectra, point-contact spectroscopy, etc.), and the role of coherent Kondo-type magnetic fluctuations is evidenced by the recovery of a conventional single-ion Kondo regime on heating up to near room temperature or, alternatively, diluting Yb by nonmagnetic Lu. The low-temperature magnetic specific heat changes from a linear gT term in the dilute limit to an exponential behavior in pure YbB_{12}, indicating that the spectrum of low-lying magnetic excitations is strongly altered.

Previous time-of-flight [1] and triple-axis [2] neutron scattering experiments on YbB_{12} had revealed that the magnetic excitation spectrum at 10 K has a gap structure with no magnetic signal below approximately 10 meV. Furthermore, a group of 3 peaks was found just near the gap edge, at 15, 20, and 38 meV. The distinct dependences of these excitations as a function of temperature and Lu substitution suggested that their origins might be different.

Experiments and results

We have carried out a detailed investigation of the magnetic spectral response in YbB_{12}. The experiments were performed on the triple-axis spectrometer 2T using an assembly of two large, high-quality, single crystals (total volume of approximately 0.4 cm^3) grown in an image furnace at Hiroshima University.

In the energy range of the measurement (0-32 meV), two branches of magnetic excitations have been observed, around 14.5 and 19 meV respectively (fig. 1), which correspond to two spectral components identified in the powder experiments. The lower mode (M1) is strongly peaked near the zone-boundary *L* point, $\boldsymbol{Q}$ = (3/2,3/2,3/2), denoting a main role of antiferromagnetic (AF) Yb–Yb correlations with the wave vector $\boldsymbol{k}$ = (1/2,1/2,1/2). This point also corresponds to a minimum in the energy dispersion (fig. 2). From spectra similar to those shown in fig. 1, we have constructed a map, in the $[1\bar{1}0]$ scattering plane, of the intensity associated with the M1 spectral component (fig. 3). It appears that this intensity is strongly reduced for $\boldsymbol{q} \parallel [110]$, and practically vanishes for $\boldsymbol{q} \parallel [001]$. Comparing experimental spectra measured at equivalent points $\boldsymbol{Q} = (q,q,q)$, for q = 1.5, 2.5, and 3.5, it was possible to estimate and subtract out the phonon component, then to trace the Q dependence of the magnetic signal M1: within experimental accuracy, its intensity does not deviate from the Yb^{3+} magnetic form factor.

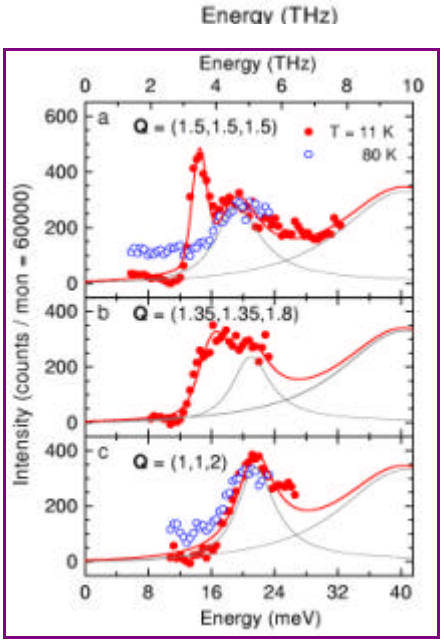

Figure 1. Magnetic neutron spectra of YbB_{12} measured for 3 different $\boldsymbol{Q}$ values on the zone boundary.

The second mode (M2) is much more extended in

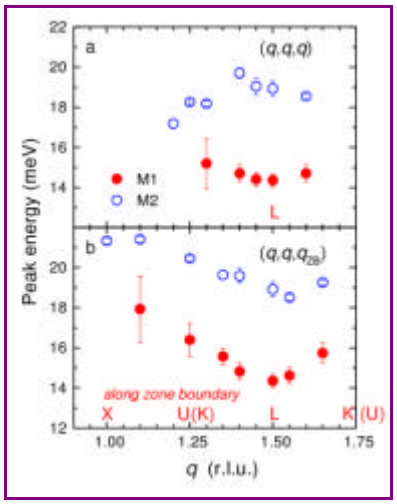

Figure 2. Energy dispersions of the two modes M1 and M2 along [111] (upper frame) and following the zone boundary (lower frame).

Q-space (compare figs. 1*a* and 1*c*). Quantitative analysis of this excitation is complicated by the fact that it is superimposed on one weakly dispersive optic phonon mode. However, phonon correction can be performed at least approximately, and the remaining signal, which decreases on going from $\boldsymbol{Q}$ = (1,1,2) to (1,1,4), is clearly magnetic in origin. Spectra measured along the [1,1,*q*] direction, where the M2 mode dominates, seem to indicate that its intensity varies across the Brillouin zone mainly because of the *q* dependence of the peak *width*. This result is surprising and may be due to an interplay between phonon and magnetic scattering. It is noteworthy that the energy of M2 exhibits significant dispersion: along the [111] direction, it decreases from zone boundary to zone center (fig. 2*a*), whereas following the zone boundary it goes through a minimum at the *L* point (fig. 2*b*).

Increasing temperature rapidly suppresses the M1 peak, and the corresponding signal becomes undetectable above 60 K. Simultaneously, a broad and *q*-independent quasielastic component is seen to grow in the spin-gap region, in agreement with the previous powder results [3] On the other hand, temperature below 80 K has no substantial effect on either the intensity or the energy dispersion of M2. The latter signal is only slightly reduced.

It can be noted that the observed *Q* dependence of the low-energy excitations explains the strong reduction of the bulk susceptibility occurring below 70 K: the reason is the transfer of magnetic spectral weight in the gap-edge region from zone center toward zone boundary.

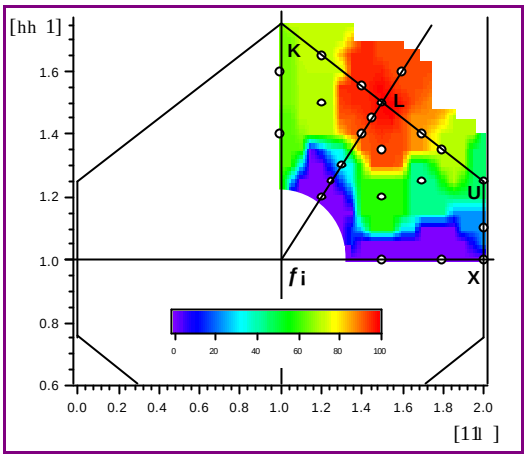

Figure 3. Interpolated intensity map of the M1 mode; circles denote *Q*-space points at which energy spectra have been measured.

Concerning the physical origin of M1, the fact that this excitation disappears completely above 60 K implies that it is intimately connected with the gap in the electronic structure, which disappears at about the same temperature, as evidenced by the optical conductivity data. The above results suggest that it originates from magnetic fluctuations at the AF *L* point. Whether the physics of these excitations should rather be treated as localized or itinerant remains an open question. Based on similarities with mixed-valence SmB_6 it is likely that the magnetic wave functions involved in the formation of AF fluctuations may be the same kind of "local bound excitonic states" (extended part of the MV wave functions) proposed to exist in the latter compound. In the present case, these states should probably involve hybridization with orbitals at neighboring Yb sites in order to explain the staggered character of the magnetic response.

As to the M2 mode, comparison with the powder results suggests that it behaves to some extent as a single-ion (e.g. crystal-field) excitation. If so, the observation of appreciable dispersion in the single-crystal spectra should be ascribed to the effect of exchange interactions, as observed many years ago by Shapiro et al. in SmS. However, a possible role of phonons needs to be clarified by polarized neutron experiments.

References

[1] A. Bouvet et al., J. Phys.: Condens. Matter 10 (1998) 5667; E.V. Nefeodova et al., Phys. Rev. B 60 (1999) 13507.
[2] Measurements on IN8 made on one of the crystals used for the present study have been reported in: F. Iga et al., J. Phys. Chem. Solids 60 (1999) 1193.
[3] It was found in the powder measurements that the upper broad peak at 38 meV also collapses above ~ 100 K.