Evidence for short-range antiferromagnetic fluctuations in Kondo insulating YbB\(_{12}\)

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The spin dynamics of mixed-valence YbB\(_{12}\) has been studied by inelastic neutron scattering on a high-quality single crystal. In the Kondo-insulating regime realized at low temperature, the spectra exhibit a spin-gap structure with two sharp, dispersive, in-gap excitations at \(\hbar \omega \approx 14.5\) and \(\approx 20\) meV. The lower mode is shown to associate with short-range correlations near the antiferromagnetic wave vector \(q_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\). Its properties are in overall agreement with those expected for a “spin exciton” branch in an indirect hybridization gap semiconductor.

Heavy-fermion compounds exhibit a whole spectrum of unconventional low-temperature behaviors, basically reflecting the existence of a very small energy scale, of the order of a few tens of kelvin, in the electron subsystem. This energy scale is a hallmark of strong electron correlations and, in the metallic case, is associated with the Kondo temperature below which the heavy-quasiparticle Fermi liquid forms. In insulating compounds, on the other hand — so-called “Kondo insulators” (KI) or “mixed-valence semiconductors” (MVSC), such as CeNiSn, Ce\(_3\)Bi\(_4\)Pt\(_3\), SmB\(_6\), YbB\(_{12}\), or UPt\(_3\) — it corresponds to the opening of a very narrow, temperature-dependent, energy gap in the electron density of states. The physical origin of this insulating state is still incompletely understood. It has been argued that a number of aspects can be explained in terms of a one-electron band picture, with a “hybridization gap” forming at low temperature in the electronic density of states at the Fermi energy. However, there is growing evidence that strong electron-electron correlations are central to the emergence of the gap behavior, and that their effects cannot be reduced to a mere renormalization of quasiparticle states. The spin dynamics of these systems is also peculiar: in most examples studied to date, inelastic neutron scattering (INS) spectra typically exhibit a spin-gap response \((\Delta_\gamma \sim 1 - 10\) meV) at low temperature, which seems directly related to the KI state and disappears rapidly when a single-site fluctuation regime is recovered by heating. Information on \(Q\) dependences has remained rather scarce, and to a large extent inconclusive, either because of complex anisotropy effects as in CeNiSn, or because measurements were carried out only on polycrystal samples. YbB\(_{12}\) is a promising candidate for further investigations: it is an archetype KI compound with a simple NaCl-type crystal structure (interpenetrating fcc sublattices of Yb ions and B\(_{12}\) cuboctahedra), and previous inelastic neutron scattering (INS) experiments on powder have indicated the presence of two narrow magnetic excitations near the spin-gap edge. Early single-crystal measurements were interpreted in terms of a single dispersive low-energy mode with high intensity along [111], but the form of the \(Q\) dependence was not clearly established. In this Letter, we report a detailed investigation of the low-energy spin dynamics in YbB\(_{12}\) showing that there indeed exist two distinct excitations, and that the lower one has a maximum in its intensity, together with a minimum in its energy dispersion, at the wavevector \(q_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\). We ascribe this behavior to short-range antiferromagnetic (AFM) correlations and argue that it could correspond to the spin-exciton branch recently predicted to occur in the KI regime of a periodic Anderson model.

The sample studied consisted of two high-quality single crystals (total volume \(\approx 0.4\) cm\(^3\)) grown by the traveling-solvent floating-zone method using an image furnace with four xenon lamps. INS experiments were performed on the thermal-beam triple-axis spectrometer 2T at the LLB (Saclay). The sample was oriented with a (110) crystal axis normal to the scattering plane, and cooled to temperatures between 10 and 80 K inside a closed-cycle refrigerator. Neutron spectra were recorded at fixed final energy, \(E_f = 14.7\) meV (PG 002 monochromator, analyzer, and filter on the scattered beam), yielding an energy resolution of \(\approx 2\) meV at zero energy transfer.

As noted in earlier powder studies, significant nuclear scattering from acoustic and lower optic phonon modes exists in the energy window of interest \(15 \leq \hbar \omega \leq 25\) meV. Special care must therefore be exerted to ensure that this signal is correctly subtracted from the experimental spectra. To this end, we have compared data obtained at several equivalent \(Q\) vectors, using lattice-dynamics measurements and numerical simulations as a guideline. The procedure is exemplified in Fig. I for \(Q = \tau + (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\), where \(\tau\) denotes a reciprocal lattice vector. Experimental data are plotted together with the calculated nuclear and magnetic components. The decomposition was made by assuming that the \(Q\) dependence of the magnetic intensity follows the Yb\(^{3+}\) atomic form factor, and that the phonon intensity varies proportional to \(Q^2\), as expected from the model calculation for this particular set of \(Q\) vectors. The important check, shown in the middle frame of Fig. I is that the
spectrum measured at the intermediate vector \((\frac{5}{2}, \frac{5}{2}, \frac{5}{2})\) is correctly reproduced using the partial (phonon and magnetic) contributions derived from the \(Q = (\frac{3}{2}, \frac{3}{2}, \frac{3}{2})\) and \((\frac{7}{2}, \frac{7}{2}, \frac{7}{2})\) spectra (Fig. 1 upper and lower frames) [27]. The contributions corresponding to the zone boundary \(X\) point, \(q = (0,0,1)\), were similarly estimated from the \((1,1,2)\) and \((1,1,4)\) spectra, yielding a phonon signal comparable to that derived above. An average of the two was thereafter taken to represent the phonon background for all measured zone boundary spectra. On the trajectory between \((\frac{5}{2}, \frac{5}{2}, \frac{5}{2})\) and the \((1,1,1)\) zone center, the phonon intensity was assumed to scale with \(Q^2\) [28]. Along \((\xi, \xi, 1)\), flat low-energy optic modes existing at about 24 to 28 meV throughout the Brillouin zone [12] hamper the separation of the magnetic contribution in this energy range.

Typical magnetic excitation spectra obtained from the above treatment are displayed in Fig. 2 for three different \(Q\) vectors on the zone boundary, \(Q = Q_0 \equiv (\frac{7}{2}, \frac{7}{2}, \frac{7}{2})\), \((1.35, 1.35, 1.8)\), and \((1, 1, 2)\), (see sketch of Brillouin zone in Fig. 3). In the upper frame, one notes a clear spin-gap region (no detectable signal below \(\approx 10\) meV) followed by two narrow peaks, \(M1\) and \(M2\), at 14.5 and 19 meV. These energies coincide with those found in the previous powder measurements, confirming that the peaks arise from the same magnetic excitations. Moving away from the \([111]\) direction, \(M1\) is strongly suppressed, while shifting to higher energies. It becomes practically undetectable in the \(c^*\) direction (bottom frame). In contrast, the \(M2\) peak remains visible for all three \(Q\) vectors, showing only a moderate increase in energy between the \(L\) and \(X\) points. The solid lines in Fig. 2 represent fits of the data using a scattering function \(S_{\text{mag}}\) constituted of three spectral components, in analogy with Refs. [8, 13].

\[
S_{\text{mag}} \propto \frac{E}{1 - \exp\left(-E/k_BT\right)} \sum_{i=1}^{3} \chi_i(Q,T)P_i(E,Q,T).
\]

The shapes of the normed spectral functions \(P_i(E,Q,T)\) were chosen Gaussian for \(M1\) (nearly resolution limited) and Lorentzian for \(M2\) as well as for the broad “40-meV” peak. The position and width of the latter component, whose maximum lies beyond the limit of our experimental energy window, were fixed (except for the form-factor \(Q\) dependence) at values obtained in the powder experiments [8]. This is justified because the excitation results primarily from single-site processes [13].

The same procedure was applied to spectra measured at a number of \(Q\) vectors, both inside and on the boundary of the \(\tau = (1,1,1)\) Brillouin zone, in order to determine the \(Q\)-dependence of the lower two excitations \(M1\) and \(M2\). The most notable result is that the intensity of \(M1\) has a pronounced maximum at the \(L\) point, which also corresponds to an absolute minimum in its dispersion curve. This is demonstrated in Fig. 8 where the energy and integrated intensity of \(M1\) are plotted as a function of the reduced \(q\) vector for trajectories through
the L point respectively parallel to [111] and along the zone boundary. The intensity drop when shifting away from \(Q_0\) is accompanied by a moderate increase in the experimental line width. An intensity map of \(M1\) covering one quadrant of the projected Brillouin zone is presented in Fig. 3. It clearly shows the maximum centered around \(Q_0\), with a steep drop in intensity on going toward the zone center. \(M1\) retains significant intensity all along \((\xi, \zeta, 1)\), possibly going through a local maximum near \(Q = (\frac{1}{2}, \frac{1}{2}, 1)\). In contrast, it is drastically suppressed along the entire \((1, 1, \zeta)\) direction.

The corresponding analysis for \(M2\) is less straightforward because of the above-mentioned overlap with low-energy optic phonons. The general trend is a weaker energy dispersion in comparison with \(M1\), and a much more uniform azimuthal distribution of the intensity. However, as in the case of \(M1\), a pronounced reduction of the integrated signal occurs near zone center. The result is unambiguous because, in this region, \(M2\) becomes narrow enough and thus can be easily separated from the phonon background. In the numerical fits, it turns out that the loss in spectral weight results mainly from a decrease in the signal line width.

With increasing temperature, one observes a complete suppression of the \(M1\) excitation (see spectra for \(T = 80\) K in Fig. 2), in agreement with the powder results [6, 7, 8]. The data also confirm the recovery of magnetic intensity, ascribed to quasi-elastic scattering, in the former spin-gap region. It should be noted that the disappearance of \(M1\) takes place without appreciable shift in the excitation energy. The \(T\) dependence of the parameters for \(M1\) and \(M2\) derived from the fit is summarized in Fig. 4 for \(Q = Q_0\). The \(M1\) peak is steeply suppressed between 25 and 60 K, with an attending increase in its line-width (left frame). On the other hand, \(M2\) (right frame) is little affected by the increase in temperature up to 80 K.

The main outcome of the present measurements is the unambiguous observation of two distinct excitations in the magnetic neutron spectrum at \(T = 10\) K, exhibiting contrasted \(Q\) and \(T\) dependences. The lower one, \(M1\), is of particular interest to us because it is specific to the low-temperature KI state. The distribution of intensity over the Brillouin zone, showing a pronounced maximum near \(Q_0 = (\frac{1}{2}, \frac{1}{2}, 1)\), analogously to an exciton in a conventional semiconductor. This view is in line with the model proposed by Riseborough in Ref. 10, which predicts a sharp branch of
in-gap dispersive “spin-exciton” excitations to form, under certain conditions, in KI systems as a result of AFM exchange interactions $J(Q)$. This process is favored by the enhancement of the real part of the dynamical susceptibility $\chi'(Q, \omega)$ taking place at the spin-gap edge for $Q$ vectors close to a zone boundary AFM point. The excitations are described as the continuation of an AFM paramagnon branch which softens at low temperature and undergoes strong narrowing due to the suppression of the electron-hole decay channel when it eventually falls below the threshold of the Stoner continuum. Moving toward the zone center, $J(Q)$ decreases and $\chi'(Q, \omega)$ broadens, leading to a positive dispersion, and the gradual disappearance, of the spin-exciton branch as observed in the present experiments.

In this interpretation, the damping, and subsequent disappearance of $M1$ with increasing $T$ is a direct consequence of the gap filling evidenced by optical and transport measurements. The model implicitly assumes that the gap forms through a hybridization process taking place coherently at each $4f$ ion site, and would thus be expected to be sensitive to disorder caused by chemical substitution on the Yb sublattice. However, neutron spectra for the Yb$_{1-x}$Lu$_x$B$_{12}$ solid solutions indicate that the spin gap is quite robust and can be traced up to 90% Lu dilution, which is at variance with its rapid suppression with increasing temperature (recovery of quasielastic scattering). This raises the interesting possibility that the spin gap could be driven by single-site effects (local spin-singlet state) as suggested by other authors, without necessarily requiring the existence of an insulating gap. Whether some degree of local character can be reconciled with the spin-exciton picture remains as a question for future investigations.

In summary, we have demonstrated a significant effect of AFM short-range correlations in YbB$_{12}$, leading to the formation of a sharp dispersive in-gap mode. The main properties of this excitation can be captured by a rather simple “spin-exciton” model, though the possibility of local effects, as inferred from substitution experiments, should also not be overlooked. Low-energy peaks have so far been reported for a few KIs such as SmB$_6$, CeNiSn, and now YbB$_{12}$, but failed to be observed in a number of others. It is noteworthy that mechanisms reminiscent of the spin-exciton formation, but based on the existence of a superconducting rather than insulating gap, have been invoked to explain anomalous magnetic modes occurring in high-$T_c$ cuprates (the well-known “resonance peak”) or the HF compound UPd$_2$Al$_3$. The picture emerging here for an archetype KI may thus prove of interest for a broader class of “spin-gap systems”.

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[27] Away from the main [111] direction, the agreement is reliable for the zone boundary $L$ point, $Q_{0} = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. As a whole, the procedure appears to be quite good for $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, somewhat less so for $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The upper mode $M2$.
[28] In Ref. 16 the effect occurs in $(\pi, \pi, \pi)$ for a simple cubic lattice, which is relevant to SmB$_6$ but not directly to YbB$_{12}$. 