

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 ≈ 20 meV. The lower mode is shown to be associated with short-range correlations near the antiferromagnetic wave vector $\mathbf{q}_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. Its properties are in overall agreement with those expected for a “spin-exciton” branch in an indirect hybridization gap semiconductor.

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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 [1]. 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,” such as CeNiSn , $\text{Ce}_3\text{Bi}_4\text{Pt}_3$, SmB_6 , YbB_{12} , or UPtSn —it corresponds to the opening of a very narrow, temperature-dependent, energy gap in the electron density of states [2]. The physical origin of this insulating state is still incompletely understood. It has been argued [3] 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 [4]. 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 ($\Delta_s \sim 1\text{--}10$ 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 [2]. Information on \mathbf{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 [5] with a simple NaCl-type crystal structure (interpenetrating fcc sublattices of Yb ions and B_{12} cuboctahedra), and previous INS experiments on powder [6–8] have indicated the presence of two narrow magnetic excitations near the spin-gap edge. Early single-crystal measurements [9] were interpreted in terms of a single dispersive low-energy mode with high intensity

along [111], but the form of the \mathbf{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 wave vector $\mathbf{q}_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. 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 [10].

The sample studied consisted of two high-quality single crystals (total volume ≈ 0.4 cm³) grown by the traveling-solvent floating-zone method using an image furnace with four xenon lamps [11]. INS experiments were performed on the thermal-beam triple-axis spectrometer 2T at the LLB (Saclay). The sample was oriented with a $\langle 110 \rangle$ 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 ≈ 2 meV at zero energy transfer.

As noted in earlier powder studies [6–8], 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 out from the experimental spectra. To this end, we have compared data obtained at several equivalent \mathbf{Q} vectors, using lattice-dynamics measurements and numerical simulations [12] as a guideline. The procedure is exemplified in Fig. 1 for $\mathbf{Q} = \boldsymbol{\tau} + (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, where $\boldsymbol{\tau}$ 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

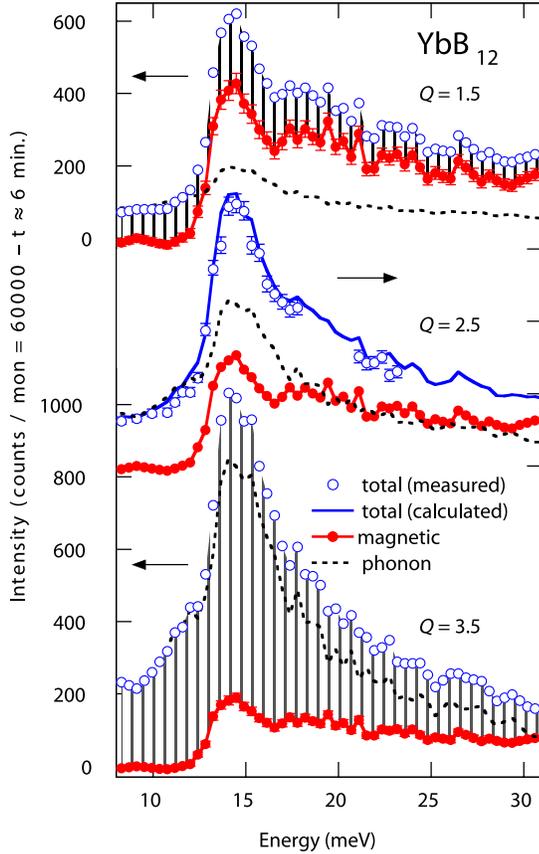


FIG. 1 (color online). Separation of nuclear and magnetic scattering (see text) in spectra measured at $T = 11$ K for three equivalent L points, $\mathbf{Q} = Q(1, 1, 1)$; hatched areas represent the phonon signal, also plotted as dashed line.

set of \mathbf{Q} vectors. The important check, shown in the middle frame of Fig. 1, is that the spectrum measured at the intermediate vector $(\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$ is correctly reproduced using the partial (phonon and magnetic) contributions derived from the $\mathbf{Q} = (\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$ and $(\frac{7}{2}, \frac{7}{2}, \frac{7}{2})$ spectra (Fig. 1, upper and lower frames) [13]. The contributions corresponding to the zone boundary X point, $\mathbf{q} = (0, 0, 1)$, 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 $(\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$ and the $(1, 1, 1)$ zone center, the phonon intensity was assumed to scale with Q^2 [14]. 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 \mathbf{Q} vectors on the zone boundary, $\mathbf{Q} = \mathbf{Q}_0 \equiv (\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$, $(1.35, 1.35, 1.8)$, and $(1, 1, 2)$ (see sketch of the Brillouin zone in Fig. 3). In the upper frame, one notes a clear spin-gap region (no detectable signal below ≈ 10 meV) followed by two narrow peaks, $M1$ and $M2$, at 14.5 and

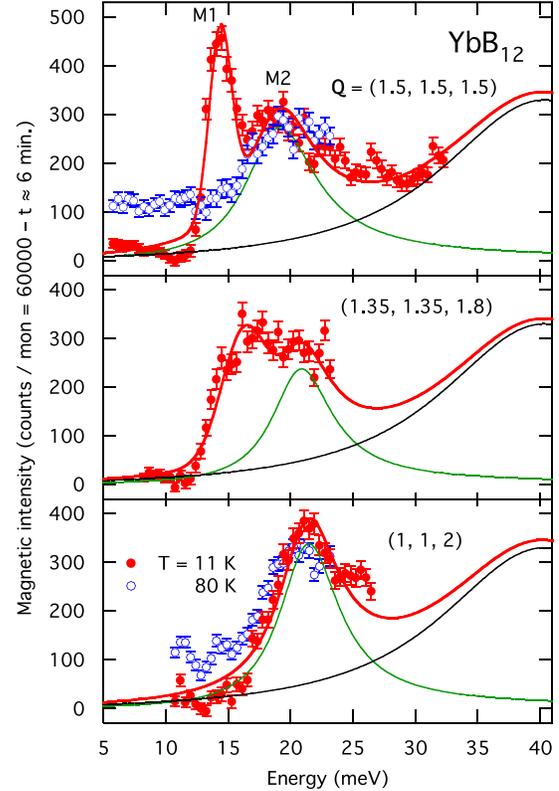


FIG. 2 (color online). Magnetic excitation spectra measured for three different scattering vectors at $T = 11$ K (closed circles) and 80 K (open circles). Solid lines represent a fit with 3 spectral components (in thin lines, $M1$ omitted for clarity).

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 \mathbf{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_{mag} constituted of three spectral components, in analogy with Refs. [8,15],

$$S_{\text{mag}} \propto \frac{E}{1 - \exp(-E/k_B T)} \sum_{i=1}^3 \chi'_i(\mathbf{Q}, T) P_i(E, \mathbf{Q}, T).$$

The shapes of the normed spectral functions $P_i(E, \mathbf{Q}, T)$ 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 \mathbf{Q} dependence) at values obtained in the powder experiments [8]. This is justified because the excitation results primarily from single-site processes [15].

The same procedure was applied to spectra measured at a number of scattering vectors, both inside and on the

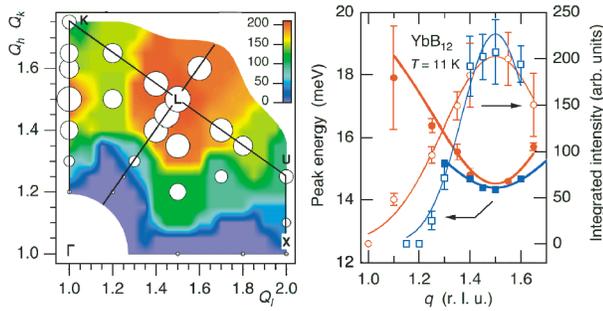


FIG. 3 (color). \mathbf{Q} dependence of the energies and intensities of the low-energy peak $M1$ at $T = 11$ K; left frame: intensity map over one quadrant of the Brillouin zone (circles denote \mathbf{Q} vectors at which energy spectra have been measured, with diameters indicative of intensities); right frame: intensities (open symbols) and dispersion curves (closed symbols) along the zone boundary (circles) and the $[111]$ direction (squares).

boundary of the $\tau = (1, 1, 1)$ Brillouin zone, in order to determine the \mathbf{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. 3, where the energy and integrated intensity of $M1$ are plotted as a function of the reduced \mathbf{q} vector for trajectories through the L point, respectively, parallel to $[111]$ and along the zone boundary. The intensity drop when shifting away from \mathbf{Q}_0 is accompanied by a moderate increase in the experimental linewidth. 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 \mathbf{Q}_0 , with a steep drop in intensity on going toward the zone center. $M1$ retains significant intensity all along $(\xi, \xi, 1)$ possibly going through a local maximum near $\mathbf{Q} = (\frac{3}{2}, \frac{3}{2}, 1)$. 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 the zone center.

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 [6–8]. The data also confirm the recovery of magnetic intensity, ascribed to quasielastic scattering, in the former spin-gap region. It should be noted that the disappearance of $M1$ takes place without an 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 $\mathbf{Q} = \mathbf{Q}_0$. The $M1$ peak is steeply suppressed between 25 and 60 K, with an attending increase in its linewidth (left frame). On the other hand, $M2$ (right frame) seems to be little affected by the increase in temperature up to 80 K.

However, neutron experiments with polarization analysis should be performed to establish the properties of $M2$ more reliably.

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 \mathbf{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 $\mathbf{Q}_0 = \tau_{111} + \mathbf{q}_0$ denotes the importance of magnetic fluctuations associated with the wave vector $\mathbf{q}_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. Short-range AFM correlations are central to the physics of metallic HF systems [16], especially near quantum critical points, and there is evidence that they play a role in KI's as well [17,18]. It has been reported [19] that some metallic heavy rare-earth RB_{12} compounds order in complex, amplitude-modulated, incommensurate structures. The paramagnetic ground state is specific to YbB_{12} , likely due to a Kondo spin-liquid-like phase [1]. The magnetic couplings responsible for the observed AFM correlations may be affected by Fermi surface changes due to the gap formation. The steep decrease in the intensity of $M1$ taking place between 25 and 60 K is directly correlated with the filling of the gap observed over the same temperature range in optical conductivity spectra $\sigma(\omega)$ [20]. Basing on a parallel between the electronic structures of YbB_{12} and metallic MV $YbAl_3$, Okamura *et al.* [21] have argued that the shoulder occurring below 40 meV in their low-temperature measurements of $\sigma(\omega)$ reflects the *indirect* gap in the renormalized density of states expected from a periodic Anderson model. In the neutron spectra, this energy of 40 meV roughly corresponds to the position of the broad peak in the time-of-flight data [8], which disappears on heating in the same temperature region ($T \approx 50$ K) as the anomaly in $\sigma(\omega)$, and we thus believe that it represents a plausible estimate of the spin-gap value. If this is right, $M1$ has to be considered an *in-gap* mode, analogous 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

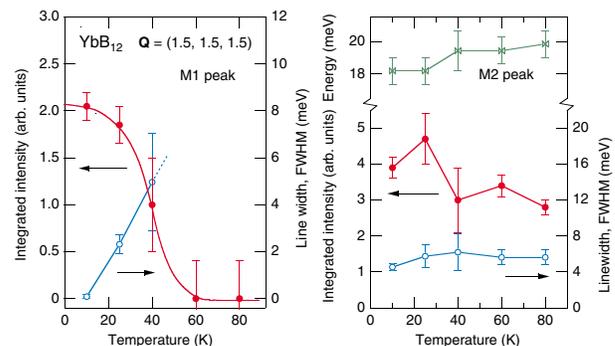


FIG. 4 (color). Temperature dependence of fit parameters for the low-energy peaks $M1$ (left) and $M2$ (right) in the energy spectra measured at $\mathbf{Q}_0 = (\frac{3}{2}, \frac{3}{2}, \frac{3}{2})$; lines are guides to the eye.

of in-gap dispersive “spin-exciton” excitations to form, under certain conditions, in KI systems as a result of AFM exchange interactions $J(\mathbf{Q})$. This process is favored by the enhancement of the real part of the dynamical susceptibility $\chi'(\mathbf{Q}, \omega)$ taking place at the spin-gap edge for \mathbf{Q} vectors close to a zone boundary AFM point [22]. 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, $J(\mathbf{Q})$ decreases and $\chi'(\mathbf{Q}, \omega)$ 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 [15] for the $\text{Yb}_{1-x}\text{Lu}_x\text{B}_{12}$ 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 [23,24], 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 [25], CeNiSn [26], and now YbB_{12} , but failed to be observed in a number of others [2]. 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”) [27] or the HF compound UPd_2Al_3 [28]. 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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- [1] H. Tsunetsugu, M. Sigrist, and K. Ueda, *Rev. Mod. Phys.* **69**, 809 (1997).
 - [2] P. S. Riseborough, *Adv. Phys.* **49**, 257 (2000).
 - [3] G. Aeppli and Z. Fisk, *Comments Condens. Matter Phys.* **16**, 155 (1992).
 - [4] G. Zwicknagl, *Adv. Phys.* **41**, 203 (1992).
 - [5] M. Kasaya *et al.*, *J. Magn. Magn. Mater.* **47–48**, 429 (1985).
 - [6] A. Bouvet, Ph.D. thesis, University of Grenoble, 1993.
 - [7] A. Bouvet *et al.*, *J. Phys. Condens. Matter* **10**, 5667 (1998).
 - [8] E. V. Nefeodova *et al.*, *Phys. Rev. B* **60**, 13 507 (1999).
 - [9] F. Iga *et al.*, *J. Phys. Chem. Solids* **60**, 1193 (1999).
 - [10] P. S. Riseborough, *J. Magn. Magn. Mater.* **226–230**, 127 (2001).
 - [11] F. Iga, N. Shimizu, and T. Takabatake, *J. Magn. Magn. Mater.* **177–181**, 337 (1998).
 - [12] K. S. Nemkovski *et al.*, *Phys. Status Solidi (c)* **1**, 3093 (2004).
 - [13] Away from the main [111] direction, the agreement is also quite good for $\mathbf{Q} = (\frac{3}{2}, \frac{3}{2}, \frac{1}{2})$ and $(\frac{1}{2}, \frac{1}{2}, \frac{5}{2})$, somewhat less so for $(\frac{1}{2}, \frac{1}{2}, \frac{3}{2})$. As a whole, the procedure appears reliable for the zone boundary L point, $\mathbf{q}_0 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.
 - [14] This rather crude approximation is sufficient here because the acoustic phonon modes are weak enough in this Brillouin zone, and their dispersion is not significant in the q range where $M1$ is observed. Only close to the zone center should one take into account an extra signal due to the bottom of the LO branch, which partly overlaps the upper mode $M2$.
 - [15] P. A. Alekseev *et al.*, *J. Phys. Condens. Matter* **16**, 2631 (2004).
 - [16] N. Grewe and F. Steglich, in *Handbook of Physics and Chemistry of Rare Earths*, edited by K. A. Gschneider and L. Eyring (Elsevier, Amsterdam, 1991), Vol. 14, p. 343.
 - [17] T. E. Mason *et al.*, *Phys. Rev. Lett.* **69**, 490 (1992).
 - [18] T. G. Rappoport, M. S. Figueira, and M. A. Continentino, *Phys. Lett. A* **264**, 497 (2000).
 - [19] I. Bat'ko *et al.*, *Appl. Phys. A: Mater. Sci. Process.* **74**, S829 (2002).
 - [20] H. Okamura *et al.*, *Phys. Rev. B* **58**, R7496 (1998).
 - [21] H. Okamura *et al.*, *J. Magn. Magn. Mater.* **272–276**, e51 (2004).
 - [22] In Ref. [10], the effect occurs in (π, π, π) for a simple cubic lattice, which is relevant to SmB_6 but not directly to YbB_{12} .
 - [23] T. Kasuya, *Europhys. Lett.* **26**, 277 (1994).
 - [24] S. H. Liu, *Phys. Rev. B* **63**, 115108 (2001).
 - [25] P. Alekseev *et al.*, *J. Phys. Condens. Matter* **7**, 289 (1995).
 - [26] T. J. Sato *et al.*, *J. Phys. Condens. Matter* **7**, 8009 (1995).
 - [27] F. Onufrieva and P. Pfeuty, *Phys. Rev. B* **65**, 054515 (2002).
 - [28] N. K. Sato *et al.*, *Nature (London)* **410**, 340 (2001).