Low-energy magnetic response and Yb valence in the Kondo insulator YbB_{12}

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X-ray absorption and high-resolution inelastic neutron-scattering experiments on the Kondo insulator YbB_{12} are reported. The ytterbium L_3-edge spectrum consists of a single white line with no observable temperature dependence between 10 and 295 K, indicating that the Yb valence is closer to 3+ than was concluded from previous high-resolution photoemission and XPS results. The neutron measurements confirm the existence of a gap in the magnetic excitation spectrum for $T=5$ K, and show that its gradual suppression as temperature increases is due to the appearance of an extra quasielastic component in the magnetic dynamical response.

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I. INTRODUCTION

Kondo insulators form an interesting class of f electron systems exhibiting a valence instability.[1] They behave as nonmagnetic narrow-gap semiconductors at low temperatures but recover a local-moment character near or above room temperature, often associated with metalliclike conductivity. SmB_6 and $Ce_3Bi_4Pt_3$ are well-known members of this family. It has been proposed that the insulating gap in their electronic density of states at the Fermi energy is due to the hybridization of electronic bands[1,2] or, alternatively, to a local Kondo interaction.[3,4] YbB_{12} is the only known Kondo insulator among Yb compounds with a valence instability. It crystallizes in the UB_{12}-type crystal structure (space group $Fm3m$). The physical properties observed near room temperature (metallic conductivity, localized magnetic moments) dramatically change when T decreases. At approximately 75 K, the paramagnetic susceptibility goes through a maximum, then decreases rapidly at lower temperatures.[5–8] A steep increase in the electrical resistivity is observed in the same temperature range.[5–7] This, together with the Schottky anomaly in the low-temperature electronic specific heat,[6,8] provides strong support to the opening of a gap in the electronic excitation spectrum. Inelastic neutron-scattering experiments[9–11] have revealed that the magnetic excitation spectrum is also anomalous, and cannot be described by conventional single-ion magnetic excitations. On the other hand, it also differs significantly from the response obtained in the other Kondo insulator $Ce_3Bi_4Pt_3$,[12] or in intermetallic MV (M=transition metal ion, V=rare earth) compounds.[13] Two important features of the magnetic response of YbB_{12} are the very-low-magnetic intensity observed at $T=15$ K for energies $E<10$ meV, and the existence of a fine structure with several distinct peaks in the inelastic spectrum just above this gaplike region. The question of the possible occurrence of quasielastic scattering at low temperature, as is generally observed in MV and heavy-fermion compounds, could not be settled by our previous study[10] because the experimental conditions (resolution of the order of 2 meV, low counting rate) did not allow such a signal to be detected if it were either concentrated in a narrow peak indistinguishable from the elastic line or, on the contrary, strongly broadened. It can be noted that recent high-resolution photoemission experiments[14,15] have detected a narrow gap of about 10 meV, which roughly corresponds to the values found in the transport measurements and in the dynamical magnetic response.

It is not yet clear whether the insulating ground state in YbB_{12} should be ascribed to a ''hybridization gap'' as predicted in mixed-valence models, or reflects the formation of a Kondo singlet on each Yb^{3+} ion. More generally, the influence of the degree of valence mixing on the formation of the Kondo-insulator state is not well documented. To shed

some light on these issues, it is important to obtain a reliable determination of the Yb valence in this material. Previous estimates from the Curie constant below room temperature yielded values of about 2.75–2.8 for polycrystals,[5,6] and 2.95 for a high-purity single crystal.[7] From the photoemission measurements,[14] the valence was estimated to be $\nu = 2.86 \pm 0.06$. Previous x-ray photoelectron spectroscopy (XPS) measurements[16] yielded $\nu = 2.9$, whereas the Yb L_3 edge x-ray absorption spectrum measured in Ref. 5 was simply denoted ''trivalent.'' Therefore, despite general agreement on the fact that the valence state of Yb is close to 3+, a more accurate determination remains desirable.

In this paper, we report x-ray absorption measurements of the Yb L_3 edge in YbB_{12} showing that the valence is indeed very close to 3+, and essentially independent of temperature. The temperature dependence of the quasielastic magnetic response, studied by high-resolution neutron-scattering experiments, is also presented and discussed in connection with the results of susceptibility measurements on the same sample. All data have been collected using the YbB_{12} powder specimen studied previously.[10]

II. EXPERIMENTS

Powder samples of $Yb^{11}B_{12}$ and $Lu^{11}B_{12}$ were prepared by borothermal reduction at 1700 °C under vacuum from elemental ^{11}B (^{11}B enrichment 99.5%) and Yb and Lu oxides, respectively. The material was characterized by x-ray diffraction. The room-temperature lattice parameters are 7.4634(2) Å for LuB_{12} and 7.4684(2) Å for YbB_{12}. The YbB_{12} sample contained traces of YbB_6 and Yb_2O_3 ($\sim$1.5%).

The x-ray absorption measurements were performed at BM12B at the BESSRC of the Advanced Photon Source of Argonne National Laboratory. The Si[111] reflection was used, yielding an energy resolution of better than 2 eV at 9 keV. Fine-ground powder was prepared, homogeneously mixed with boron nitrate, and pressed into a pellet, the thickness of which was chosen so that $\mu x \leqslant 1$ (μ: absorption coefficient; x: sample thickness). This pellet was attached to the cold finger of a closed-cycle refrigerator and cooled down to temperatures of the order of 10 K. Energy calibration was obtained using data for a trivalent Yb standard (Yb_2O_3), which were collected simultaneously.

The inelastic neutron-scattering experiments were carried out on the time-of-flight spectrometer IN6 at the Institute Laue Langevin (ILL) in Grenoble. The incident neutron energy was $E_0 = 3.14$ meV, yielding a resolution of $\Delta E = 100\,\mu$eV at zero energy transfer. Polycrystalline samples of YbB_{12} (7.5 g) and LuB_{12} (5.4 g) were filled into flat aluminum containers with dimensions $23 \times 45 \times 1.9\,mm^3$ and $23 \times 45 \times 1.5\,mm^3$, respectively. In order to subtract the background contribution from the sample environment, an empty sample holder and a sample holder containing a cadmium plate instead of the sample were also measured. The spectrum of a vanadium standard was used for absolute calibration. Because of the low incoming neutron energy, the momentum transfer remains small ($\sim$1.5 $Å^{-1}$) over the scattering angle range $10° < 2\theta < 150°$. Measurements were carried out at temperatures $T = 5$, 35, 50, 80, 120 K, and $T = 5$, 80, 120 K for YbB_{12} and LuB_{12}, respectively. Typical measuring time for one spectrum was approximately 5 h.

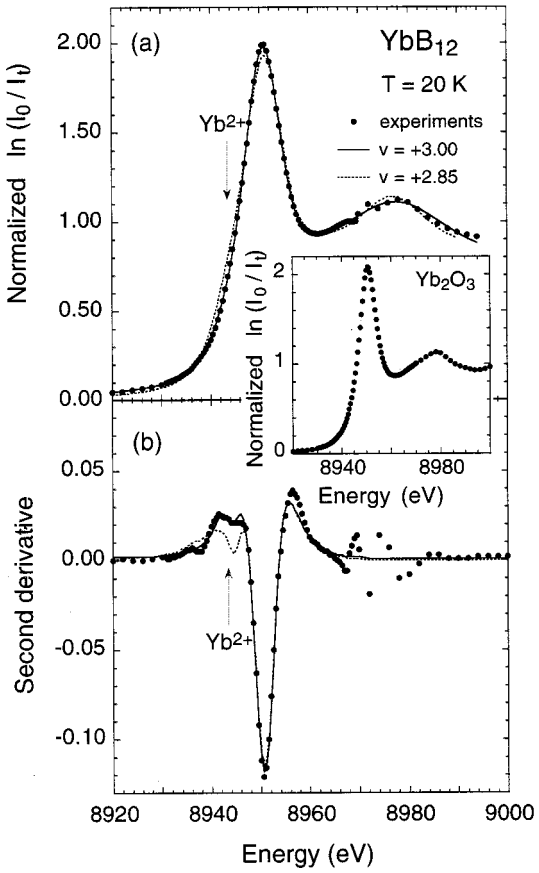

FIG. 1. X-ray absorption spectrum of Yb in YbB_{12} measured at $T = 20$ K (a) and its second derivative (b); solid (dotted) lines are fits assuming an average Yb valence $\nu = 3$ ($\nu = 2.85$). Inset: measured Yb L_3 absorption edge for the trivalent reference compound Yb_2O_3. I_0 and I_t correspond to the x-ray intensity of the incoming and transmitted beam, respectively.

The susceptibility measurements were performed using a commercial (Quantum Design) physical properties measurement system (PPMS) device.

III. RESULTS

The x-ray absorption spectrum taken at $T = 20$ K is shown in Fig. 1(a), together with a smoothed plot of the second derivative in Fig. 1(b). The second derivative of the x-ray absorption near-edge structure (XANES) spectrum is commonly used to visualize smaller contributions to the edge data, in particular for systems exhibiting noninteger valence states. Here the XANES spectrum exhibits a single resonance at $E = 8949$ eV similar to that found in Yb_2O_3 (inset in Fig. 1), where Yb is unambiguously trivalent. The data are well represented by an *arctan* function and a Lorentzian, as it is often the case for x-ray absorption spectra at the L_3 edges of trivalent rare-earth ions.[17] A fit to the data yields a Lorentzian half-width of 4.2 ± 0.3 eV, in good agreement with the calculated natural line width of the core hole (3.9 eV).[18] In

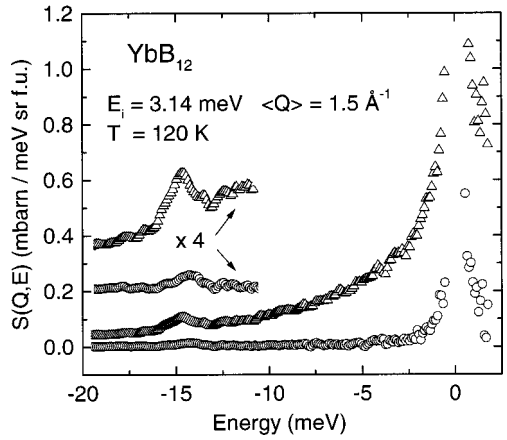

FIG. 2. Inelastic neutron-scattering spectrum for YbB_{12} (△) and LuB_{12} (○) at $T=120$ K, averaged over scattering angles between 20° and 60°. The data in the range $-20<E<-10$ meV are also shown on an expanded scale (×4), with the origin shifted by 0.2 along the vertical axis.

Fig. 1(b), one can see that the second derivative of the XANES spectrum is well described taking into account only purely trivalent Yb. On the contrary, a fit assuming a valence of 2.85 (dotted lines) fails to reproduce the experimental data on the low-energy side of the peak, where a contribution from 15% Yb^{2+} should be easily detected. Finally, we have found that the L_3-edge data at $T=10$ and 295 K are indistinguishable in the near-edge region within experimental accuracy.

The neutron time-of-flight spectra for YbB_{12} and LuB_{12} at $T=120$ K are shown in Fig. 2. The data were corrected for self-screening, taking into account the geometry of the neutron experiment. To improve statistics, all spectra corresponding to scattering angles lower than that of the first Bragg peak ($20°\leqslant 2\theta\leqslant 60°$) have been added up. Data points in the range $-0.5\leqslant E\leqslant 0.5$ meV have been discarded because the signal was strongly contaminated by elastic scattering. The YbB_{12} spectrum at $T=120$ K exhibits extra intensity in the energy range $-20.0\leqslant E\leqslant 2.5$ meV in comparison with that of LuB_{12}, which is used as a reference system with only nuclear scattering. The YbB_{12} and LuB_{12} spectra both contain one distinct inelastic peak at $E\sim -15$ meV, which has been previously attributed to the phonon density of states.[10] By comparing the spectra for the two compounds, it can be concluded that the YbB_{12} spectrum contains a broad, structureless magnetic component. This component can be singled out by subtracting the inelastic and elastic nuclear scattering defined from the LuB_{12} data measured at the same temperature ($T=5$, 80, and 120 K). For temperatures of 35 and 50 K, at which no reference spectra were available, the nuclear contribution was estimated on the basis of the LuB_{12} spectrum at $T=120$ K, taking into account the detailed-balance factor in the case of the phonon component.

The magnetic response $S_{\mathrm{mag}}(E)$ of YbB_{12} at $T=120$ K is depicted in Fig. 3(a). It cannot be fitted by a single spectral component but is well described by two—one broad and one narrow—quasielastic Lorentzian functions with $\Gamma_1/2=12$ meV and $\Gamma_2/2=0.8$ meV. With decreasing temperature,

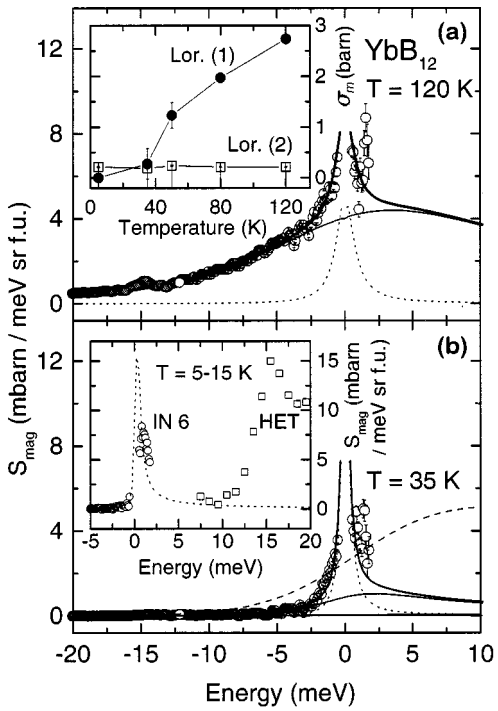

FIG. 3. Magnetic excitation spectrum of YbB_{12} at $T=120$ K (a) and 35 K (b). Thick lines are fits to a two-component spectral function consisting of one broad (solid line) and one narrow (dashed line) Lorentzians (see text and Table I). The broken line in frame (b) represents the broad component calculated for $T=35$ K assuming the same line width ($\Gamma/2=12$ meV) and magnetic cross section as at $T=120$ K. Inset in frame (a): integrated intensities (detail balance corrected) of the broad (1) and narrow (2) quasielastic components as a function of temperature. Inset in frame (b): magnetic response of YbB_{12} measured on IN6 (ILL) at $T=5$ K (○) and on HET (ISIS) (Ref. 10) at $T=15$ K(□).

the intensity of the broad component is gradually suppressed [Fig. 3(b)]. At the lowest experimental temperature of 5 K, only the narrow quasielastic component can be observed [inset in Fig. 3(b)] because of the detailed-balance factor and limited accessible energy range on the energy-loss side. The parameters of the two Lorentzian functions for different temperatures are collected in Table I. The integrated intensities (in absolute units and corrected with detailed balance) representing the magnetic cross section σ_m of the narrow and broad components in the magnetic spectral function are shown in the inset of Fig. 3(a) as a function of temperature. For the broad component, there is a clear increase of the intensity with temperature, which seems especially pronounced in the temperature range around 50 K. The narrow component, on the other hand, does not show any significant temperature dependence.

The strong suppression of the broad quasielastic component below 50 K is illustrated by the dashed line in Fig. 3(b), which represents the contribution calculated from the data at 120 K assuming that the signal remains quasielastic and that the total cross section does not change with temperature (i.e., only the variation of the detailed-balance factor is taken into account). Obviously, the result strongly overestimates the contribution found experimentally at $T=35$ K. The inset in

TABLE I. Fitting parameters of the broad (1) and narrow (2) quasielastic components in the magnetic response of YbB_{12}.

	Lorentzian (1)		Lorentzian (2)	
Temperature (K)	Half-width (meV)	Integrated intensity (barn)	Half-width (meV)	Integrated intensity (barn)
5			0.40 ± 0.15	0.20 ± 0.1
35	5 ± 3	0.3 ± 0.3	0.40 ± 0.1	0.19 ± 0.05
50	11.0 ± 1	1.2 ± 0.3	0.50 ± 0.15	0.20 ± 0.1
80	10.4 ± 0.4	1.97 ± 0.08	0.60 ± 0.15	0.2 ± 0.1
120	11.8 ± 0.4	2.74 ± 0.08	0.80 ± 0.1	0.22 ± 0.1

Fig. 3(b) shows a combination of the data from Ref. 10 and from the present measurements, which demonstrates the existence of a gaplike region below $E=10$ meV. The narrow quasielastic component will be discussed more specifically in the following.

IV. DISCUSSION

The existence of a single resonance in the L_3-edge spectrum of YbB_{12} indicates that Yb is essentially trivalent. Any extra divalent contribution would lead to a resonance centered 7 eV lower in energy as found, e.g., in the MV compound $YbInCu_2$.[19] In particular, the spectrum calculated for a valence of 2.85 (roughly one Yb^{2+} for six Yb^{3+}), as deduced from the high-resolution photoemission[14] experiments, does not appear to represent our data correctly. The introduction of a smaller Yb^{2+} contribution does not produce any significant improvement over the single-line fit, and we estimate the upper limit of the Yb^{2+} fraction in YbB_{12} to be no more than 5% ($\nu \geqslant 2.95$). Similarly, the fit of the second derivative in Fig. 1(b), assuming purely trivalent Yb, does not reveal any additional divalent contribution to the absorption spectrum. From the measurements at $T=10$ K, it can be concluded that the valence of Yb in YbB_{12} does not change significantly as a function of temperature.

One possible origin for the difference between our results and previous photoemission studies may be the different sensitivities of these methods to surface effects. Whereas the measurement of the Yb L_3 edge at $E=8944$ eV is a bulk probe sampling a depth of approximately 20 μm, the XPS and the high-resolution photoemission measurements are much more surface sensitive. In principle, photoemission can differentiate between the surface (the upper layer of the material) and the ''bulk'' (the next few layers) from the surface shift due to the different potentials existing at the surface and in the bulk. The above-mentioned high-resolution photoemission data[15] clearly showed that the Yb at the surface is divalent. It is not unlikely that the valence in the next layer may also be somewhat reduced as the *f* electrons are located close to the Fermi surface. It is also possible that defects close to or at the surface may influence the Yb valence of the near-surface layers probed by photoemission. For the XPS measurements, the electron escape depth is in the order of 20 Å, corresponding to roughly five layers of Yb ($a/2=3.7$ Å between adjacent Yb layers). Assuming a divalent surface layer of Yb as observed in the high-resolution photoemission experiment, this contribution is in the order of 15%–20%, which would indicate a bulk Yb valence of very close to $\nu=3$ in good agreement with our estimate from the hard x-ray absorption at the L_3 edge. We therefore believe that the difference between our results and the high-resolution photoemission and XPS data is due to the surface sensitivity of the XPS and photoemission techniques.

The main result of the inelastic neutron study is the unambiguous confirmation that a nonmagnetic singlet ground state, with a gap in the magnetic excitation spectrum, exists in YbB_{12} at $T=5$ K. The formation of this gap is associated with the suppression of the broad quasielastic contribution observed at higher temperatures, which becomes particularly steep below 50 K. This spectral component is observable only above 35 K but, as soon as it appears, it shows a quite significant line width ($\Gamma/2\approx5$ meV). Accordingly, the closing of the gap with increasing temperature does not result from a shift of excitation threshold to lower energies but rather from the emergence of the extra quasielastic signal.

It is interesting to compare these results with the low-temperature magnetic susceptibility χ for the same sample, which is plotted in Fig. 4. Below room temperature, $\chi(T)$ first decreases on cooling, then goes through a maximum

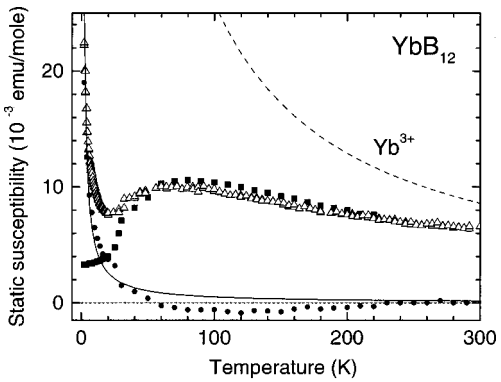

FIG. 4. Temperature dependence of the magnetic susceptibility of YbB_{12}, measured on polycrystal (△: this work) and single-crystal (■: from Ref. 8) specimens. Dashed line: Curie law calculated for a free Yb^{3+} ion (magnetic moment $\mu=4.54\mu_B$). Closed circles: ''impurity'' contribution obtained as the difference between the polycrystal and single-crystal data. Solid line: fit of the latter contribution to a single-ion Curie law assuming 2% of Yb^{3+}.

around 75 K, in agreement with previous polycrystal[5,6] and single-crystal measurements.[7,8] The decrease of $\chi(T)$ below the maximum is consistent with the temperature dependence of the magnetic response, in particular with the disappearance of the broad quasielastic component in approximately the same temperature range. On the other hand, the observation of a Curie tail below 12 K is most likely an impurity effect because a similar feature was observed previously in polycrystalline samples,[5,6] but is almost negligible for a high-quality single crystal.[7,8] The same magnetic impurities are most likely responsible for the narrow quasielastic peak observed in all our neutron spectra. The magnetic cross section obtained by integrating the intensity over this peak at $T=5$ K is 0.21 b. This value corresponds to approximately 2% of the total magnetic cross section, $\sigma_m=12.6$ b, calculated for Yb^{3+}. By fitting the ''impurity'' tail in $\chi(T)$ by a Curie law with the effective magnetic moment of Yb^{3+} ($p_{\text{eff}}=4.54\mu_B$), one obtains an impurity concentration again of the order of 2%. It should be noted that this extra term does not necessarily reflect the existence of an impurity *phase* in the chemical sense (although this possibility cannot be ruled out within the accuracy of our x-ray characterization of the material): indeed, it has been reported recently[20] that if powder is produced by crushing the single-crystal material of Ref. 7, its magnetic susceptibility exhibits a low-temperature tail like that of polycrystals.

The existence of a broad structureless magnetic response, and the formation of a gap in the magnetic excitation spectrum below $\approx$50 K have been reported previously for the Kondo insulator $Ce_3Bi_4Pt_3$.[12] In a recent paper, Liu[21] proposed to ascribe the magnetic gap in the latter system to the formation of an array of local nonmagnetic singlet states, resulting from the hybridization and Coulomb interaction between localized ($4f$) and band ($5d$) electrons. A similar approach could be applicable to YbB_{12}. According to that model, the thermal breakdown of the local nonmagnetic bound state should result in the growth of the quasielastic contribution due to the population of the excited magnetic Yb^{3+} states hybridized with the conduction band. Consequently, one expects to recover a spin-fluctuation spectrum corresponding to Kondo effect on Yb^{3+} ions at elevated temperatures, which could explain the origin of the broad quasielastic response in YbB_{12} for $T>50$ K. Further theoretical work along this line seems quite promising.

In conclusion, the x-ray absorption study of YbB_{12} has shown that Yb is essentially trivalent in the bulk ($\nu \geqslant 2.95$), and that its valence does not change significantly with temperature. The Kondo-insulator ground state of YbB_{12} can be characterized as nonmagnetic, with a gap in the magnetic excitation spectrum for temperatures below 35 K. At higher temperatures, a broad quasielastic contribution starts to appear in the magnetic response, providing evidence that the Yb ions cross over to a spin-fluctuation regime with a fluctuation energy of the order of 10 meV in this temperature range. The fact that the appearance of a magnetic quasielastic intensity goes hand in hand with the closing of the electronic gap suggests that a strong correlation exists between the form of the magnetic response and the electronic band structure in this material. Finally, it should be noted that, in contrast to other compounds such as SmB_6 or $Ce_3Pt_4Bi_3$, the Kondo-insulator gap state in YbB_{12} does not involve a significant admixture of two different valence states.

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