An Inelastic Neutron Scattering Study of the Spin Dynamics of \( \text{Yb}_{1-x}\text{Lu}_x\text{Al}_3 \)

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An Inelastic Neutron Scattering Study of the Spin Dynamics of Yb$_{1-x}$Lu$_x$Al$_3$

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We present the results of a systematic inelastic neutron scattering study of the spin dynamics of the mixed valent compound YbAl$_3$ doped with nonmagnetic lutetium. The aim of the investigation is to clarify the origin of the unusual gap–like magnetic response observed in YbAl$_3$, which can be modeled by two inelastic peaks: a narrow peak at 34 meV with HWHM, $\Gamma = 6.4 \pm 0.8$ meV and a broad peak at 44 meV with $\Gamma = 30 \pm 1$ meV. Lutetium substitution leads to a substantial increase in the linewidth ($\Gamma = 9 \pm 1$ meV at $x = 0.1$) and a decrease in the intensity (down by 60% at $x = 0.1$) of the narrow component, with a negligible effect on the broad inelastic peak. This trend is confirmed with higher doping resulting in the complete suppression of the narrow peak at $x \geq 0.35$. The results indicate that the narrow component arises from coherent excitation processes within the hybridized 4f$^7$-band, which are destroyed by disorder, while the broad component is not so sensitive to the loss of coherence.

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I. INTRODUCTION

The intermetallic compound YbAl₃ has been the subject of numerous investigations into its mixed valent (MV) behavior, which is due to the hybridization of the localized 4f electrons with the conduction electrons [1-5]. Inelastic neutron scattering (INS) experiments, which directly probe the imaginary part of the dynamic magnetic susceptibility, have revealed an unusual gap-like spectral response at low temperature [6]. This is characterized by a very weak level of magnetic scattering at low energies (less than 30 meV), followed by a sharp threshold at 32 meV with a broad inelastic response above this value [6-8]. This has been ascribed both to excitations across a hybridization gap in the f-band at low temperature [6], or to the results of an electron-phonon interaction [8]. There have also been claims, on the basis of cold-neutron experiments, that the gap-like response does not exist at all [9].

In an attempt to clarify the origin of this magnetic response, we have performed a systematic INS investigation of its evolution in Yb₁₋ₓLuₓAl₃ as a function of lutetium concentration and temperature. Doping with lutetium does not produce significant chemical pressure (as in the case of Y or La dilution) owing to tiny (~0.3%) differences in the lattice parameters of YbAl₃ and LuAl₃. Therefore, we expect that the main effect of the substitution of lutetium for ytterbium on the magnetic response of Yb₁₋ₓLuₓAl₃ will be through the disruption of periodicity in the ytterbium sublattice. This should allow us to distinguish between collective and single ionic aspects of the spectral response.
II. EXPERIMENT

The samples of $Yb_{1-x}Lu_xAl_3$, with $x = 0.0, 0.05, 0.1, 0.25,$ and $0.35$, were prepared by arc-melting of the constituent elements with $\sim5\%$ excess of Yb because of the high vapor pressure of ytterbium. The weight loss after the melting procedure did not exceed 4-5%. Both neutron and x-ray diffraction confirmed that the samples were single phase. The sample masses with $x = 0.0$ and 0.1 were $\sim50g$ and those with $x = 0.05, 0.25,$ and 0.35 were $\sim10g$. The $Yb_{0.5}Lu_{0.5}Al_3$ sample ($\sim30g$) was the same as in the Ref. [6]. The INS measurements were performed at the pulsed spallation neutron source ISIS (Rutherford Appleton Laboratory, U.K.) on the time-of-flight chopper spectrometer HET, using incident energies of 20 and 150 meV with FWHM resolutions at the elastic positions of 0.23 and 2.0 meV, respectively. The samples were sealed in thin aluminium sachets and mounted on a closed-cycle refrigerator for scans down to 12K. The data were calibrated on an absolute scale by comparison with a standard vanadium sample.

III. DATA ANALYSIS

A. Correction for Phonon Scattering

The main challenge in the quantitative interpretation of measured INS spectra is to derive the phonon contribution correctly, particularly in compounds, such as $YbAl_3$, that have a strong nuclear cross section. In our view, the safest method is usually the one first described in Ref. 10, in which a non–magnetic reference compound is used to define how the phonon scattering (single + multiple) scales with scattering angle (see Ref. 11 for a detailed review of this technique). The magnetic compound's own high–angle scattering, where the magnetic intensity is weakest, can then be used to estimate the low–angle phonon background.
Unfortunately, LuAl₃ is not a reliable non-magnetic reference because of the large absorption cross section of lutetium, which is associated with a resonance at 140 meV. Therefore, we have used a modification of this technique that permitted the scaling function between low and high-angle phonon scattering to be estimated without the use of an isostructural nonmagnetic compound. We have made two assumptions. Firstly, we assumed that the spectral response of YbₓLu₁₋ₓAl₃ at high temperatures (T > 150 K) is purely quasielastic and has a Lorentzian line shape in accord with previous INS studies of YbAl₃ [6,7,9]. Secondly, we assumed that the scaling function between low and high-angle phonon scattering can be represented by a simple linear function Aω + B, where ω is the energy transfer and A > B. This form of the scaling function has been experimentally established for numerous compounds measured on HET and is justified by computer simulations [11]. Having made these two assumptions, we can write the following equation:

\[ S(\phi_L, \omega) = S_{mag}(\phi_L, \omega) + (A \omega + B) \left( S(\phi_H, \omega) - S_{mag}(\phi_L, \omega) \left( \frac{f^2(Q(\phi_H, \omega))}{f^2(Q(\phi_L, \omega))} \right) \right) \]  

where the magnetic contribution \( S_{mag}(\phi_L, \omega) \) is given by

\[ S_{mag}(\phi_L, \omega) = f^2(Q(\phi_L, \omega)) \frac{\omega \chi_0}{(1 - \exp(-\omega/kT)) \left[ \omega^2 + \Gamma^2 \right]} \] 

\( S(\phi_L, \omega) \) and \( S(\phi_H, \omega) \) are the experimentally measured scattering laws at low and high scattering angles, \( f(Q(\phi_L, \omega)) \) is the Yb⁺³ magnetic form factor, \( \chi_0 \) is the static susceptibility, and \( \Gamma \) the half-width at half-maximum (HWHM) of the Lorentzian. The last term in Eqn. 1 is a correction for the small magnetic contribution to the high-angle data, which is especially important for experiments with low incident energies. We estimated the values of the four adjustable parameters, \( \chi_0, \Gamma, A, \) and \( B \), by fitting the high
temperature data to Eqn. 1 convoluted with the spectrometer resolution function, and then kept $A$ and $B$ fixed when analyzing the lower temperature data.

B. Analysis of Magnetic Scattering

Figure 1 shows data (open circles) from YbAl$_3$ measured at an average angle $\langle \phi \rangle = 19^\circ$ with an incident energy of 20 meV at 300, 200, 150, and 50 K. After subtracting the phonon contribution (dotted lines), estimated by the method discussed in the previous section, the magnetic scattering is seen to be broad and structureless over the energy range $-50$ to 15 meV, and decreases with temperature. The purely relaxational spectral function (Eqn. 2) provides an excellent description of the measured spectra. Both the linewidth of the spectral response and the static susceptibility, $\chi_0$, are nearly temperature-independent for $T > 150$ K ($\Gamma = 24.2 \pm 0.4$ meV, $\chi_0 = (4.1 \pm 1.0) \times 10^{-3}$ emu/mol at 300 K). The susceptibility is in good agreement with the bulk measurements ($\chi_0 = 4.5 \times 10^{-3}$ emu/mol at 300 K [1]), which show a broad maximum at around 130 K. This behavior is typical of mixed valent systems. A similar relaxational response is seen in the lutetium-doped compounds with the parameters shown in Table 1.

Having determined the phonon scaling function at high temperatures, we can estimate the nonmagnetic contribution to the low-angle data at low temperatures ($T<150$K). As can be seen in Fig. 2, the magnetic intensity at $T = 12$ K below $\sim 30$meV has a very low level and practically all of the magnetic spectral weight is situated at energies above $\sim 30$meV. These findings indicate that the pure relaxational model is no longer a valid representation of the spin dynamics at these temperatures. In order to characterize quantitatively the measured spectra at low temperature, a least-squares analysis was done, assuming the phenomenological model of $S_{\text{mag}}(\phi_L, \omega)$ to be the sum of
two inelastic peaks. A narrow Lorentzian centered at $\Delta_1 = 34$ meV and a broad Lorentzian centered at $\Delta_2 = 44$ meV. The narrow peak models the gap-like threshold observed in earlier studies, while the broad peak models the high-energy tail as well as the weak response seen below the threshold. The solid lines in Fig. 2 show that a two-component model fits the experimental data accurately for $x = 0.0$ and 0.1.

The increase in lutetium concentration has a strong effect on the parameters of the spectral response, especially on the narrow peak at $\sim 34$ meV. This peak gradually disappears with increasing $x$ and, at $x > 0.25$, the spectral response can be characterized by a single inelastic peak (Table 2). The broad inelastic peak has a much weaker dependence on lutetium concentration and remains practically unchanged up to $x = 0.35$, i.e., in the same concentration range where the narrow inelastic feature is still visible. In the case of $x = 0.5$, we found that the inelastic peak shifted to $69 \pm 2$ meV and broadened substantially. The total static susceptibility is independent of $x$ within the experimental error.

IV. SUMMARY AND DISCUSSION

We have presented the results of a systematic neutron scattering study of the spin dynamics of the mixed valent compounds Yb$_{1-x}$Lu$_x$Al$_3$ as a function of lutetium concentration and temperature over an energy range up to 140 meV. The main features of the observed magnetic response can be summarized as follows:

1) At high temperatures ($T > 150$K), the spin dynamics of Yb$_{1-x}$Lu$_x$Al$_3$ exhibit a quasiehstic Lorentzian form with a nearly temperature-independent linewidth.

2) With decreasing temperature, the quasiehstic spectral intensity of Yb$_{1-x}$Lu$_x$Al$_3$ is transferred progressively to an inelastic response. In the case of YbAl$_3$, the magnetic
response at 12K can be described by two inelastic peaks, a narrow Lorentzian centered at 34meV and a broad Lorentzian centered at 44meV.

3) The low temperature spectral response of Yb_{1-x}Lu_{x}Al_{3} has a strong evolution with lutetium concentration. The 34 meV peak falls dramatically with doping whereas the 44 meV peak is nearly unaffected by doping.

The broad peak is similar to the inelastic peaks seen at low temperature in other mixed valent compounds [12–15]. For example, its energy, \( \Delta = 44 \pm 3 \text{ meV} = 4 kT_{\text{max}} \) where \( T_{\text{max}} \) the maximum of the bulk susceptibility (\( T_{\text{max}} = 130 \text{ K} \) [1]). This is in good agreement with the Fermi liquid relations established in other MV compounds [14]. The parameters characterizing this peak are only weakly dependent on the level of lutetium doping for \( x \leq 0.35 \). Above this value, i.e. when only one broad Lorentzian is needed to fit the data, the peak broadens substantially and shifts to higher energy (\( \Delta = 69 \pm 2 \text{ meV} \) when \( x = 0.5 \)). The origin of this increase is not understood, but in other respects, the broad scattering is consistent with single-impurity models of mixed valent behavior.

On the other hand, the narrow component is strongly dependent on the doping level. Since the estimated phonon contribution is nearly identical for both \( x = 0.0 \) and 0.1, the sharp decrease in intensity seen in Fig. 2 is unlikely to be due to an electron–phonon coupling, as discussed in Ref. 7. It is also unlikely to result from the effect of chemical pressure since the lattice parameters of the two parent compounds, YbAl_{3} and LuAl_{3} differ by only \( \sim 0.3\% \) (\( a_{0} = 4.203 \text{ Å} \) and 4.190 Å respectively). Finally, it cannot be due to a localized crystal field excitation, since the Yb^{3+} ion should have a Kramers doublet ground state that would give rise to quasielastic scattering at low temperature. This is not observed.
The most likely explanation is that the narrow peak depends on the coherence of the hybridization of the f-electrons with the conduction electrons. Models of the scattering from coherent hybridized bands contain a high-frequency \( Q \)-dependent structure superimposed on a \( Q \)-independent background [16]. The \( Q \)-dependent component of \( S(Q,\omega) \) is associated with excitation across a hybridization gap. Although detailed comparisons are not possible due to the absence of microscopic calculations of the electronic structure of \( \text{YbAl}_3 \), these predictions are in qualitative agreement with our experimental findings. This would suggest that the origin of the gap is similar to those seen in Kondo insulators such as \( \text{Ce}_3\text{Bi}_4\text{Pt}_3 \) [17]. However, in the case of \( \text{YbAl}_3 \) there is no transport gap, presumably because of the existence of non-hybridized bands at the Fermi level. Further insight into the origin of the gap-like component of the magnetic response of \( \text{YbAl}_3 \) will require INS experiments on single crystal samples [18].

**ACKNOWLEDGMENTS**

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REFERENCES

TABLES

Table 1

Parameters of the magnetic response of Yb_{1-x}Lu_xAl_3 at T = 200K measured with an incident energy of 150meV

<table>
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<tr>
<th>$x$</th>
<th>0.0</th>
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<th>0.35</th>
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<td>$\Gamma$ (meV)</td>
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<td>30(1)</td>
<td>32(4)</td>
<td>33(2)</td>
<td>35(3)</td>
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<tr>
<td>$\chi_0$ ($\times 10^3$ emu/mol)</td>
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<td>4.6(1)</td>
<td>4.7(2)</td>
<td>3.9(1)</td>
<td>3.1(1)</td>
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Table 2

Parameters of the magnetic response of Yb_{1-x}Lu_{x}Al_{3} at T = 12K measured with an incident energy of 150meV

<table>
<thead>
<tr>
<th>x</th>
<th>0.0</th>
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<td>Δ₁ (meV)</td>
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<td>45(4)</td>
<td>46(5)</td>
<td>47(4)</td>
<td>42(2)</td>
<td>69(2)</td>
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<tr>
<td>Γ₁ (meV)</td>
<td>30(1)</td>
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<td>31(4)</td>
<td>32(4)</td>
<td>39(4)</td>
<td>45(4)</td>
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<tr>
<td>χ₁ (x 10⁻³ emu/mol)</td>
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<td>2.8(3)</td>
<td>2.6(3)</td>
<td>2.9(3)</td>
<td>3.0(3)</td>
<td>3.4(2)</td>
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<tr>
<td>Δ₂ (meV)</td>
<td>33.8(3)</td>
<td>35.3(7)</td>
<td>37(1)</td>
<td>36(2)</td>
<td>—</td>
<td>—</td>
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<tr>
<td>Γ₂ (meV)</td>
<td>6.4(8)</td>
<td>8(2)</td>
<td>9(1)</td>
<td>8(4)</td>
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<td>χ₂ (x 10⁻³ emu/mol)</td>
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<td>0.8(2)</td>
<td>0.6(2)</td>
<td>0.4(3)</td>
<td>—</td>
<td>—</td>
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<tr>
<td>χ_{total} (x 10⁻³ emu/mol)</td>
<td>3.5(4)</td>
<td>3.6(5)</td>
<td>3.2(5)</td>
<td>3.3(6)</td>
<td>3.0(3)</td>
<td>3.4(2)</td>
</tr>
</tbody>
</table>
FIGURE CAPTIONS

Figure 1

Neutron inelastic scattering from YbAl$_3$ v temperature at an average scattering angle of 19° measured with incident energy of 20meV. The solid line represents the results of fitting the data with the scattering function given by Eqs. 1 and 2. The dotted line is the estimated phonon contribution. The dashed line is the pure magnetic contribution.

Figure 2

Neutron inelastic scattering from YbAl$_3$ and Yb$_{0.9}$Lu$_{0.1}$Al$_3$ at an average scattering angle of 5° measured with incident energy of 150meV at 12K. The lines have the same meanings as in the Fig. 1. The parameters of the fit are listed in Table 2.
Figure 1
Figure 2

$S(Q, \omega)$ [mb/meV/sr/μ] vs. Energy transfer [meV] for YbAl$_3$ and Yb$_{0.9}$Lu$_{0.1}$Al$_3$.