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NEUTRON INELASTIC SCATTERING EXPERIMENTS ON THE
MIXED-VALENT COMPOUND YbCuAl

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ABSTRACT

The dynamical susceptibility of YbCuAl has been established by means of time of flight neutron scattering experiments. Non-Korringa behaviour of the quasi-elastic line-width has been found and comparison with NMR data will be given.

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1. Introduction

In order to obtain microscopic information on the dynamical behaviour of the Yb ions at temperatures both above and below the gradual valence transition in YbCuAl, we have performed inelastic neutron scattering experiments at the Institut Laue-Langevin (Grenoble, France). To separate expected phonon contributions to the scattering from the magnetic part, we also performed experiments on the non-magnetic analogue YCuAl. The YbCuAl and YCuAl specimens (about 50 g each) have been prepared in closed Mo crucibles, as described earlier [1]. YbCuAl and YCuAl are isostructural, ordered intermetallic compounds, crystallizing in the hexagonal Fe_2P type structure [2,3], in which the RE component occupies equivalent sites of low symmetry. The gradual valence transition of the Yb ions occurs in a very accessible temperature range: below 30 K the system enters a regime where the mixed-valent state exhibits a Fermi-liquid behaviour, whereas above 30 K a more localized character is observed [1]. Therefore we have performed neutron inelastic scattering experiments at 5, 30, 100 and 250 K.

2. Experimental results

Experiments have been carried out by means of the time of flight spectrometer IN4, with incident neutron energies of 12 and 50 meV and at scattering angles between 9° and 160° . Vanadium was used as a standard and background and self-shielding corrections were applied.

The spectra taken with 50 meV incident neutrons show a measurable intensity up to energy transfers of about 40 meV (fig.1). The peaks centered at 10 and 19 meV which are also present in YCuAl, can be attributed to the phonon density of states. A comparison of the spectra shows that there is also a broad magnetic contribution to the total scattering from YbCuAl. To extract the magnetic contribution from the spectra, the following procedure has been used: at high scattering angles ($2\theta = 154.1^\circ$), both the YbCuAl and YCuAl spectra were found to have the same temperature dependence. This means that there is hardly any magnetic scattering at these angles, as should also be expected from the negligible 4f form factor at these Q-values. We have fitted the high angle data with two Lorentzians with fixed positions and half-width values. The integrated intensities of these two phonon peaks were found to be nearly temperature independent, apart from their Bose factor. The problem of subtracting the correct phonon contributions at small scattering angles was satisfactorily solved by scaling down the high angle phonon contribution in YbCuAl, assuming the same Q-dependence as in YCuAl. The remaining intensity of the YbCuAl spectra has been fitted as a quasi-elastic contribution, which was assumed to have a Lorentzian shape centered at $\omega = 0$. The sum of the phonon and quasi-elastic contributions gives good fits to the spectra (fig.1). For

low Q-values the measured intensities of the quasi-elastic Lorentzians are equal to the measured static susceptibilities.

3. Discussion

Since no line-width data are available for normal magnetic trivalent RECuAl compounds, we have no reference with which to compare the measured values of $\Gamma/2$ (fig.2). We find the YbCuAl line-width large in the sense that $\Gamma/2 \gg k_B T$ for $T \lesssim 30$ K. Such large line-widths at low temperatures are invariably observed in mixed-valent systems [4,5]. Compared with most mixed valent systems studied so far, the $T_{\max}$, i.e. the temperature of the broad maximum in the susceptibility, is fairly low (~ 27 K) for YbCuAl. Our results therefore indicate a slow increase of the line-width up to $T \sim 10 T_{\max}$, the highest temperature covered in the present measurements.

Additionally, the spectral-width at 5 K is found to be larger than that at 30 K suggesting that the line-width goes through a minimum around $T_{\max}$, in agreement with the nuclear relaxation data reported earlier [6], see fig.2. It is important, however, to point out that our phonon subtraction procedure appears to slightly underestimate the phonon contribution at the lowest temperature of 5 K, suggesting perhaps that additional inelastic contributions are present, which apparently grow with decreasing temperature. These could have substantial influence on the resulting half-widths from the fits, especially at the lowest temperature of 5 K. Thus the present neutron data cannot be interpreted as firmly establishing the novel feature of the minimum in the line-width around $T_{\max}$, but as merely lending nominal support to the observations of such a temperature dependence from the nuclear relaxation measurements [6].

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Figure Captions

Fig.1. Double differential cross section versus neutron energy transfer for YbCuAl at 5 K ($2\theta = 12.4^\circ$ and 154.1°), 30 K and 100 K (both $2\theta = 12.4^\circ$). The plus signs correspond to the observed scattering.

Fig.2. Line-widths of YbCuAl as a function of temperature.