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1998 J. Phys.: Condens. Matter 10 9123

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Copper and indium Knight shifts above and below first-order valence transition in YbInCu_4

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Received 27 March 1998

Abstract. ^{63}Cu and ^{115}In Knight shifts were measured for YbInCu_4 both above and below the first-order valence transition temperature T_v . The hyperfine coupling constants of ^{63}Cu , which are small, of the order of the classical lattice dipole field, increase markedly below T_v , while that of ^{115}In exhibits an only slight change at T_v , suggesting different roles of the ligands in the valence transition.

1. Introduction

A number of investigations of YbInCu_4 , which shows a first-order valence transition at around $T_v = 40$ K, have already been accumulated in order to elucidate the origin of the *first-order* transition [1]. The 4f electrons behave as local moments above T_v and mixed with conduction bands to form a Fermi liquid state below T_v . Since non-metallic properties, say, semimetallic bare-conduction bands [2], are suggested for YbInCu_4 , we intend to detect microscopically the locally different conduction electron (hole) distribution in the crystal or anisotropic interaction of Yb 4f electrons with different ligands. The hyperfine coupling constant deduced from the Knight shift gives such local information at each ligand site. So far, however, results of only ^{115}In NMR have been reported [3–5]. The deduction of the Knight shift at the Cu site from an NMR spectrum needs a more complicated procedure due to the presence of the electric quadrupole interaction at the lower symmetric site, and hence needs well resolved experimental data of an ideally crystallized specimen. Recently, Kinder *et al* succeeded in growing a single crystal of YbInCu_4 with much better quality than earlier polycrystalline samples [6]. In the present study, we measured the temperature dependence of Knight shifts at the Cu site as well as the In site using such a well crystallized sample.

2. Experimental procedures

A crystal of YbInCu_4 was grown by the same procedure described in [6]. For NMR measurements the crystal was crushed to fine powder with diameter smaller than $50\ \mu\text{m}$. Spin-echo measurements were made with a home-made phase-coherent pulsed spectrometer both above and below T_v . YbInCu_4 forms the cubic C15b-type crystal structure (space group $F\bar{4}3m$). The site symmetry of the In site (4c site) is cubic while that of the Cu site

(16d site) is threefold around the $\langle 111 \rangle$ axis. Thus the Cu nucleus (nuclear spin $I = 3/2$ for ^{63}Cu) interacts with the electric field gradient with the asymmetry parameter $\eta = 0$. For analyses, gyromagnetic ratios $\gamma/2\pi = 1.1285$ and 0.93295 MHz kOe $^{-1}$ are used for ^{63}Cu and ^{115}In , respectively.

3. Experimental results

Figure 1 shows an example of the ^{63}Cu NMR spectrum, where only the central line (transitions between $m = -1/2$ and $+1/2$ levels) is shown. One can see a sharp and well resolved structure characterized mainly by the second-order quadrupole interaction and slightly by the anisotropic Knight shift. (The pure quadrupole frequency ν_Q is 14–15 MHz for ^{63}Cu [7].) A step corresponding to the singularity at $\theta = 0^\circ$ (θ : the angle between the local symmetry axis and the external field), which is often smeared out for an inhomogeneous sample, can be clearly seen. At each temperature, we estimated isotropic and anisotropic Knight shifts by applying both analytic calculation using singular-angle field values and computer simulation to reproduce the whole line shape. Both methods give practically the same results. In the analyses, we fixed quadrupole frequencies to the experimental zero-field values obtained by NQR measurements [7]. The temperature dependence of the Knight shifts was observable below T_v as well as above T_v . The estimated isotropic and axial Knight shifts K_{iso} and K_{ax} defined as $K(\theta) = K_{iso} + K_{ax}(3\cos^2\theta - 1)$ are plotted against the susceptibility [6] with temperature as an implicit parameter in figure 2. Points fall on different straight lines above and below T_v . Slopes of the lines give isotropic and anisotropic hyperfine coupling constants, $A_{iso} = N\mu_B K_{iso}/\chi$ and $A_{aniso} = 2N\mu_B K_{ax}/\chi$, as shown in table 1.

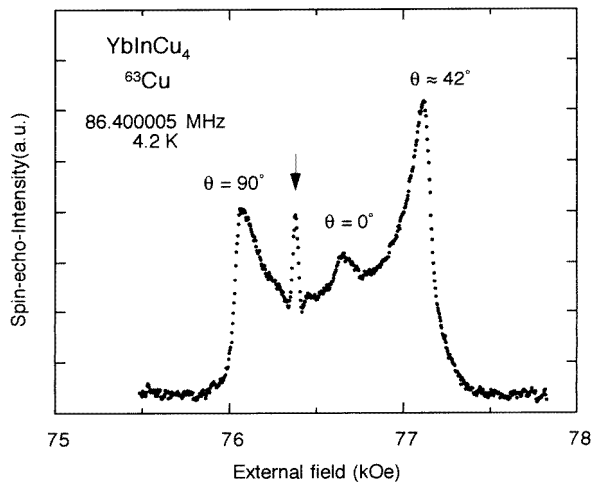


Figure 1. The centre line (transitions between $m = +1/2$ and $-1/2$ levels) of the ^{63}Cu NMR spectrum of YbInCu_4 measured at 4.2 K and 86.400005 MHz. The sharp peak indicated by an arrow is ascribed to pure metallic Cu in the pick-up coil.

We have also measured ^{115}In spectra, which have roughly symmetric Lorentzian shapes. From the peak position, we estimated the Knight shifts, which are plotted against the susceptibility in figure 3. Hyperfine coupling constants for the ^{115}In site are also presented in table 1. The value above T_v agrees well with those previously reported, while that below

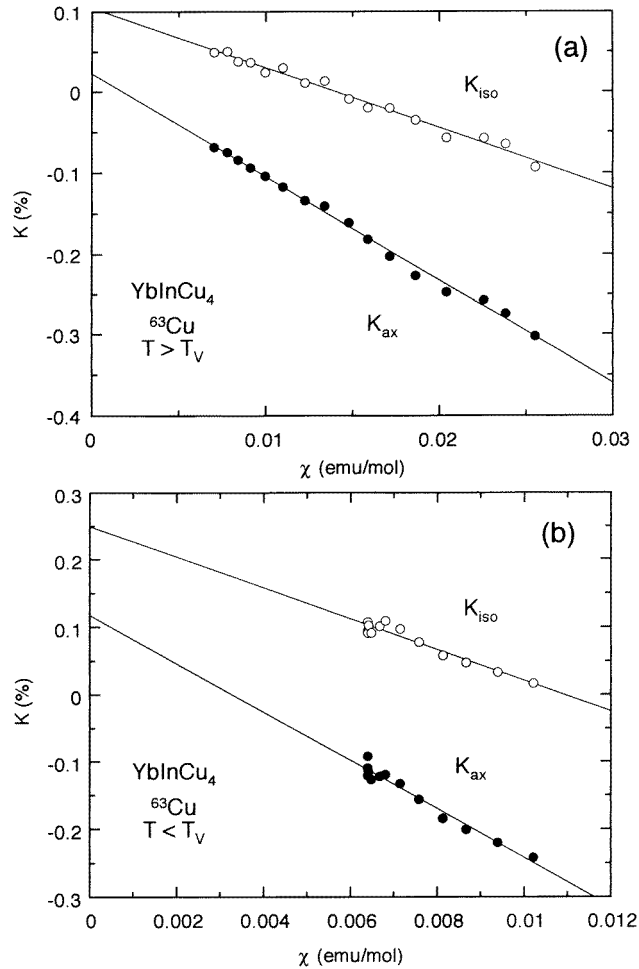


Figure 2. Isotropic (open circles) and axial (solid circles) Knight shifts of ^{63}Cu in YbInCu_4 (defined as $K(\theta) = K_{\text{iso}} + K_{\text{ax}}(3 \cos^2 \theta - 1)$) plotted against the susceptibility for above T_v (a) and below T_v (b). Slopes of straight lines give hyperfine coupling constants listed in table 1.

Table 1. Hyperfine coupling constants for YbInCu_4 .

	Cu		In
	A_{iso} (kOe μ_B^{-1})	A_{aniso} (kOe μ_B^{-1})	A_{iso} (kOe μ_B^{-1})
$T < T_v$	-1.3	-4.0	-2.5
$T > T_v$	-0.4	-1.4	-3.5

T_v is estimated to be somewhat larger than earlier results [3–5]. Although the hyperfine coupling constants below T_v may include large errors due to the possible overestimation of the temperature variation of the susceptibility, the estimated absolute value is considered to give the lower limit. This is the same for the Cu site.

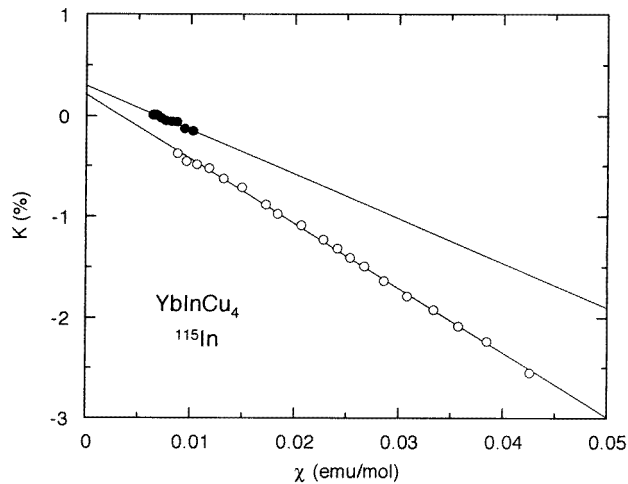


Figure 3. Knight shifts of ^{115}In in YbInCu_4 plotted against the susceptibility. Open and solid circles indicate the data above and below T_v , respectively. Slopes of straight lines give hyperfine coupling constants listed in table 1.

4. Discussion

Absolute values of both isotropic and anisotropic hyperfine coupling constants of the Cu site increase by a factor of three below T_N . Usually we expect a larger contact (isotropic) hyperfine field in the hybridized state, which is in accordance with the experimental observation for the Cu site. It is of interest to consider the origin of the anisotropy of the hyperfine interaction. The lattice sum of the dipolar field produced by Yb moments gives a calculated anisotropic coupling constant $A_{\text{aniso}}(\text{dip}) \approx -0.68 \text{ kOe } \mu_B^{-1}$. Above T_v the experimental isotropic field is practically negligible and the anisotropic term is only enhanced by a factor of two from the calculation, indicating substantially no coupling between Cu and Yb 4f electrons above T_v in accordance with the semimetallic character of this compound. Below T_v , the sudden occurrence of the hybridization gives rise to an increase of the hyperfine field at the Cu site. It should be noted that the anisotropic contribution is much larger than the isotropic one. On the other hand, the hyperfine field at the In site does not modified markedly at T_v and is even smaller below T_v than above T_v . These results suggest that the electron mixing is strongly anisotropic and that the electronic state around only the Cu site is sensitively affected by the valence transition. This argument is in qualitative agreement with the results of nuclear spin–lattice relaxation rate $1/T_1$; the discontinuous change of $1/T_1$ at T_v is larger for the Cu site [7] than the In site [5]. Hence we conclude that the 4f ligand interaction is strongly anisotropic, pointing to the non-metallic character, and that the vicinity of the electronic state to a gap or semimetallic state may be responsible for the first-order valence transition.

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