Transformation of the magnetic state in $Yb_{0.5}In_{0.5}Cu_2$ in a first-order phase transition

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The intermetallic compound $Yb_{0.5}In_{0.5}Cu_2$ doped with Gd^{3+} atoms is investigated using ESR and measurements of the static magnetic susceptibility. The evolution of the magnetic state both below and above the first-order phase transition, which is observed at 50 K in this compound, is studied. It is established that at low temperatures $Yb_{0.5}In_{0.5}Cu_2$ is a dense Kondo lattice, which passes to a regime with well defined localized magnetic moments for the Yb^{3+} ions as the temperature rises. The phase transition is associated with a Kondo volume collapse mechanism. The values of the RKKY exchange integral J_{Gd-Yb} and the spin fluctuation rate τ^{-1} of the ytterbium ions are determined. © *1996 American Institute of Physics.* [S1063-7761(96)02204-4]

1. INTRODUCTION

In the past decade the electronic properties of mixed-valence systems and heavy fermions have attracted the unwavering attention of investigators. This interest is due to the observation of behavior which is anomalous from the standpoint of classical metals and semiconductors. One member of this class of compounds is $Yb_{0.5}In_{0.5}Cu_2$. It is remarkable that this compound exhibits abrupt changes in its physical properties, viz., its static magnetic susceptibility, specific heat, lattice constants, etc., which are associated with the occurrence of a first-order phase transition, at $T\sim 50$ K.[1–4]

The compound $Yb_{0.5}In_{0.5}Cu_2$ has been actively investigated by various experimental techniques, and the following findings have been obtained.

1. A first-order phase transition occurs at $T_p=50$ K.

2. Above T_p the Yb ions behave like a system of $Yb^{3+}(4f^{13},{}^2F_{7/2})$ magnetic states, while below T_p they behave like the nonmagnetic Yb^{2+} singlet.

3. At $T<T_p$ the values of the static magnetic susceptibility $\chi(0)=0.0045$ emu/mole and the specific-heat factor $\gamma(0)=56$ mJ/mole·K are anomalously large for ordinary metals.

4. The phase transition is accompanied by only a slight change in valence, from 2.8 to 2.9.

The properties of $Yb_{0.5}In_{0.5}Cu_2$ were interpreted using the phenomenological model proposed by Felner and Nowik.[1] This model presupposes that the phase transition is associated with an abrupt change in the valence of the ytterbium ions from Yb^{2+} at low temperatures to Tb^{3+} at high temperatures. There is strong feedback between the excitation energy E_1, which separates the Yb^{2+} and Yb^{3+} states, and the occupation number p_3 in the form $E_1=E_0(1-\alpha p_3)$, where E_0 is the ground-state energy. From this standpoint $Yb_{0.5}In_{0.5}Cu_2$, is a mixed-valence compound with a small admixture of Yb^{3+} ions to Yb^{2+} ions below the phase transition and, conversely, with an admixture of Yb^{2+} ions to Yb^{3+} ions above T_p. The Felner–Nowik model provides satisfactory agreement with the experimentally observed temperature dependence of the magnetic susceptibility, specific heat, and magnetization in strong magnetic fields.[1,3,4] However, the observed slight change in valence from 2.8 to 2.9 (Ref. 3) is at sharp variance with this model. To ascertain the microscopic picture of the phase transition in $Yb_{0.5}In_{0.5}Cu_2$ we undertook a study of this material using electron spin resonance (ESR) and measurements of the magnetic susceptibility. It is known that ESR has been used successfully to study heavy fermions and variable-valence compounds (see, for example, Refs. 5–8).

We investigated the temperature dependence of the linewidth $\Delta H(T)$ and the g-factor shift $\Delta g(T)$ of paramagnetic Gd^{3+} ions implanted in $Yb_{0.5}In_{0.5}Cu_2$.

It was discovered that at low temperatures there is an intense source of spin relaxation, which practically precludes the existence of the nonmagnetic divalent states of the ytterbium ions. At the same time, the ESR linewidth increases with the temperature according to a Korringa law with a large slope, which is characteristic of heavy-fermion

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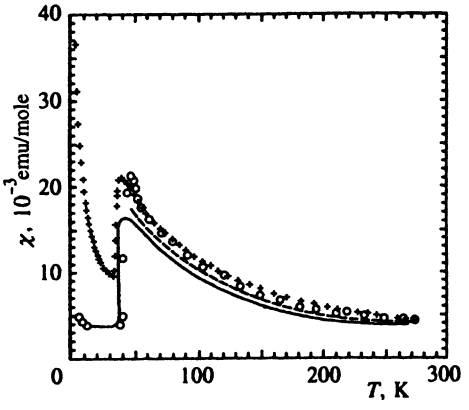

FIG. 1. Temperature dependence of the static magnetic susceptibility: O—undoped sample of $Yb_{0.5}In_{0.5}Cu_2$ +—sample of $(Yb_{0.5}In_{0.5})_{0.98}Gd_{0.02}Cu_2$. The solid line shows the dependence of $\chi(T)$ in $(Yb_{0.5}In_{0.5})_{0.98}Gd_{0.02}Cu_2$ minus the susceptibility of the Gd^{3+} ions. The dashed line shows the results of calculations using Eq. (3).

materials.[8,9] Taking into account this fact, as well as the above properties,[2–4] we proposed that at $T<T_p$ the system is in a Kondo regime with a Kondo temperature T_K greatly exceeding the crystal-field splitting Δ of the Yb ions. At $T=T_p$ there is a transition from the state of a dense Kondo lattice to a state with well defined localized magnetic moments for the Yb^{3+} ions. It is perfectly likely that the nature of the phase transition has much in common with the mechanism of Kondo volume collapse predicted by Allen and Martin.[10]

To analyze the experimental data on $\Delta H(T)$ and $\Delta g(T)$ we used the expressions proposed by Coldea *et al.*.[7] for heavy-fermion systems. Good agreement with experiment was obtained.

The values of the constants obtained from the analysis of the experimental data, i.e., the Kondo temperature T_K, the RKKY exchange integral J_{Yb-Gd}, and the spin correlation rate τ^{-1} of the Yb ions, were perfectly reasonable. Our results account for the behavior of the system over the entire range of temperatures and confirm our proposed model.

2. EXPERIMENTAL METHOD AND RESULTS

The measurements of the static magnetic susceptibility were performed at $1.5<T<300$ K for two samples: $Yb_{0.5}In_{0.5}Cu_2$ with a residual content of gadolinium ions equal to 0.01 at. % and a sample doped with 2 at. % Gd^{3+}. Figure 1 presents the results of the investigation. Both samples exhibit a sharp drop in the susceptibility at $T\simeq 40$ K. Our results are qualitatively consistent with the data in Refs. 1–4. The quantitative disparities are probably due to the method used to prepare the samples. The sample containing 2% Gd exhibits a gradual increase in the static magnetic susceptibility at low temperatures, which can be attributed to the contribution of the Gd^{3+} ions.

The ESR measurements were performed on a Varian E-101 spectrometer at 9.3 GHz in the temperature range $1.5<T<300$ K. An Oxford Instruments continuous-flow He cryostat was used for $T>4.2K$, and an He cold-finger bath cryostat was employed for $T<4.2$ K. The measurements

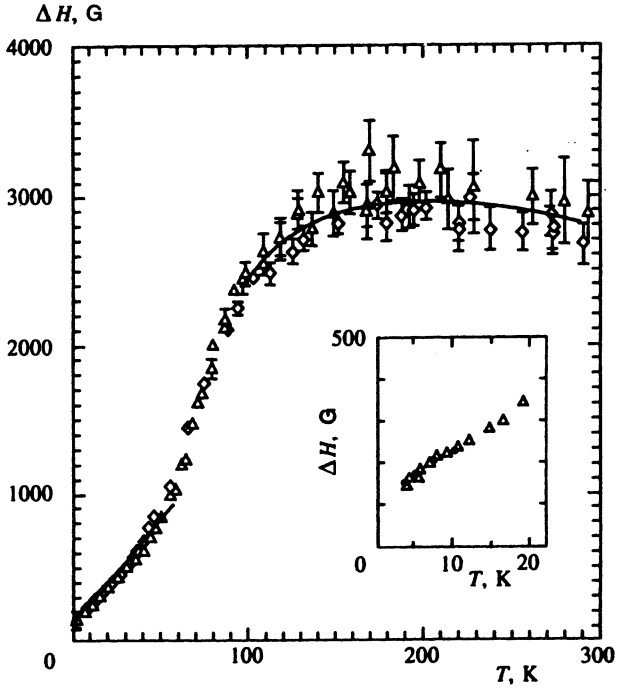

FIG. 2. Temperature dependence of the linewidth $\Delta H(T)$ for $Yb_{0.5}In_{0.5}Cu_2$ doped with 0.01 at. % Gd (●), 2 at. % Gd (△), and 5 at. % Gd (◇). All the curves have been normalized to a single residual linewidth at $T=0$. The solid line shows the results of calculations using Eq. (13).

were performed on three samples containing 0.01, 2, and 5 at. % Gd. All the samples were ground and dispersed in paraffin oil. The experimental results are shown in Figs. 2–4.

It is noteworthy that the plots of the temperature dependence of the linewidth $\Delta H(T)$ for all three samples coincide, while the plots of $\Delta g(T)$ differ significantly. The residual linewidths for the samples with 2 at. % and 5 at. % Gd differ by 100 Oe. However, in Fig. 2 we neglected this fact in order to demonstrate the absence of a dependence of the plots of $\Delta H(T)$ on the concentration of Gd^{3+} ions. The ESR linewidth increases linearly with the temperature below T_p, a sharp increase in $\Delta H(T)$ is observed in the region of the phase transition, and $\Delta H(T)$ achieves saturation above T_p. The phase transition is smoothed somewhat in comparison with the picture observed when the magnetic susceptibility was measured.

The measurements of the temperature dependence of the g-factor showed that, unlike the plots of $\Delta H(T)$, the course of $\Delta g(T)$ is highly dependent on the concentration of Gd^{3+} ions. Figure 3 shows that abrupt variation of the g-factor is observed for the sample with 2 at. % Gd^{3+} in the region of the phase transition at $T\simeq 60$ K. The phase transition is blurred in the case of the sample with 5 at. % Gd^{3+} (see Fig. 4). The linewidth is very large for $T>T_p$, and the error in the determination of the g factor is therefore strongly increased.

3. ANALYSIS OF THE EXPERIMENTAL RESULTS

A. Static magnetic susceptibility

Figure 1 presents the results of the measurements of the static magnetic susceptibility for two samples of

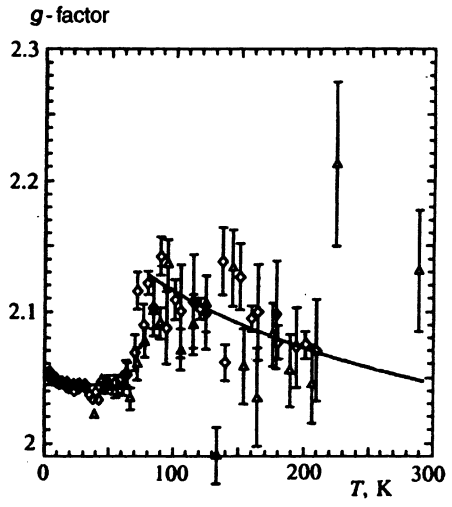

FIG. 3. Temperature dependence of the g factor of $Yb_{0.5}In_{0.5}Cu_2$ doped with 2 at. % Gd. The solid line shows the results of calculations using Eq. (12).

$Yb_{0.5}In_{0.5}Cu_2$, an undoped sample and a sample containing 2% Gd. The observed disparity between the plots of $\chi(T)$ is clearly due to the contribution of the Gd^{3+} ions. This contribution can be calculated from the formula

$$\chi_{Gd}=y\frac{N_0 g^2\mu_B^2}{3k_BT}S(S+1), \tag{1}$$

where N_0 is Avogadro's number, μ_B is the Bohr magneton, g is the spectroscopic splitting factor of the Gd^{3+} ion, $S=7/2$ is the spin of the Gd^{3+} ion, and y is the atomic fraction of the gadolinium atoms.

The calculation showed that the plots of the temperature dependence of the magnetic susceptibility for the two samples almost coincide, except in the region of the phase transition (Fig. 1). The lowering of the maximum of $\chi(T)$ for the doped sample cannot be attributed to a simple change in the number of Yb atoms (by 2%). This finding is related to the replacement of Yb^{3+} ions located at lattice sites by Gd^{3+} ions. The ionic radius of Gd^{3+} is 10% greater than that of Yb^{3+} (0.938 and 0.858 Å). It was shown in Ref. 3 that the phase-transition temperature depends on the radius of the impurity ion. The larger is its radius, the greater is the shift of the phase-transition point toward higher temperatures.

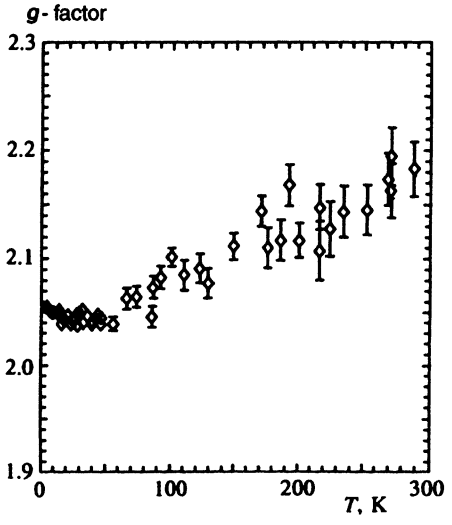

FIG. 4. Temperature dependence of the g-factor of $Yb_{0.5}In_{0.5}Cu_2$ doped with 5 at. % Gd.

This situation can also be interpreted as an effect of a "negative pressure" on the phase transition.[3] Our evaluation shows that the lattice deformation associated with the replacement of ytterbium ions by gadolinium ions displaces the phase-transition temperature by 10 K, if only the first coordination sphere is taken into account. The total perturbed part of the doped sample is equal to mxy, where $m=4$ is the coordination number of the Yb sites (we take into account only the ytterbium ions) and x is the atomic fraction of the ytterbium atoms. The electronic state of the Yb ions in the perturbed volume clearly changes to Yb^{3+} at a higher temperature than in the remainder of the sample, and the susceptibility maximum at the phase transition should be lower than in the undoped sample by $\approx mxy$, in good agreement with experiment.

As is seen from Fig. 1, for $T>50$ K the susceptibility increases with decreasing temperature, undergoes an abrupt drop at $T=T_p$, and at low temperatures is constant and equal to $\chi(T)=0.0042$ emu/mole. This value is much greater than the susceptibility usually observed in materials containing divalent Yb^{2+} ions. In addition, as has already been noted, the valence of ytterbium scarcely varies at the phase-transition point and is close to +3. Therefore, we had to seek another model for the phase transition, instead of the Felner–Nowik model. The existence of a dense Kondo state with a characteristic energy significantly greater than the crystal-field splitting was postulated at low temperatures. Rajan[11] proposed an expression for the static susceptibility of a system with $S>1/2$ in the Kondo regime:

$$\chi=x\frac{N_0\nu(\nu^2-1)\mu_B^2 g_f^2}{24\pi k_B T_K}. \tag{2}$$

Here $\nu=2J+1$ is the multiplicity of the Yb^{3+} ions, $J=7/2$, $g_f=8/7$ for Yb^{3+} ions, and $\chi=0.0042$ emu/mole (Fig. 1).

Using (2), we can evaluate the Kondo temperature for $Yb_{0.5}In_{0.5}Cu_2$ We obtained $T_K\sim 389$ K. The total splitting of the cubic crystal field for $Yb_{0.5}In_{0.5}Cu_2$ is $\Delta\sim 135$ K (Ref. 1). Thus, $\Delta\ll T_K$. These evaluations support our proposed model. If $T_K>\Delta$ holds, the static susceptibility χ clearly does not depend on the temperature below the phase-transition point.

As the temperature rises, the system passes from the highly correlated regime to a state with well defined, independent localized magnetic moments for the Yb^{3+} ions. The static magnetic susceptibility of the Yb^{3+} ions in the regime of localized states ($T>50$ K) is a sum of two components:

$$\chi=\chi_r+\chi_p, \tag{3}$$

where χ_r is the orientational part, which is associated with the variation of the populations of the energy levels and equals

$$\chi_r = C_0 x \frac{2(7/27) + 4(65/189)\exp(-E_8/T) + 2(3/7)\exp(-E_7/T)}{Z(T+\Theta)}, \tag{4}$$

and χ_p is the polarization part of the same nature as the Van Vleck contribution

$$\chi_p = C_0 x \frac{T}{\Delta} \frac{2(320/81) - 4(3008/2835)\exp(-E_8/T) - 2(64/35)\exp(-E_7/T)}{Z(T+\Theta)}. \tag{5}$$

Here $Z = 2 + 4\exp(-E_8/T) + 2\exp(-E_7/T)$; $C_0 = N_0 g_f^2 J(J+1)/3k_B$ is the Curie constant of the Yb^{3+} ions; Θ is the Curie–Weiss constant.

As is seen from (4) and (5), the splitting of the ground-state octet of the Yb^{3+} ion by the cubic crystal field of the Laves phase was taken into account in the calculations of χ_r and χ_p: the octet splits into a ground-state Γ_6 doublet, an excited Γ_8 quartet, and an upper Γ_7 doublet. If Δ is the total splitting, then $E_8 = 3\Delta/8$ and $F_7 = \Delta$ (we assumed that the energy of the lower Γ_6 doublet equals zero, and the energy levels are measured in temperature units). All the parameters were taken from Ref. 1, i.e.,

$$E_6 = 0\text{K}, \qquad E_8 = 50\text{K}, \qquad E_7 = 135\text{K}, \qquad \Theta = 20\text{K}.$$

It is seen from Fig. 1, which presents the results of the calculations, that good agreement with experiment was obtained.

B. ESR measurements

As is seen from Figs. 2 and 3, ESR, like the other methods of investigation, "senses" the phase transition in $Yb_{0.5}In_{0.5}Cu_2$ A comparison of the temperature dependences of the ESR characteristics and the static magnetic susceptibility χ in $Yb_{0.5}In_{0.5}Cu_2$ reveals that the g-factor shift Δg mimics the course of the magnetic susceptibility, but the feature in the temperature dependence of χ corresponding to the phase-transition point is shifted toward higher temperatures.

The ESR characteristics in metals are usually described by the indirect exchange interaction of impurity ions with lattice ions through conduction electrons (the RKKY interaction). Owing to the short-range character of RKKY exchange, the Gd and Yb ions experience only the influence of their local environment. Thus, the deformation of the electronic shell of a Yb ion near a Gd ion has a far stronger influence on the ESR characteristics than on the total static susceptibility of the sample and is the cause of the apparent displacement of the phase-transition point toward higher temperatures.

Unlike the samples containing 2 at. % Gd, the picture of the phase transition in $Yb_{0.5}In_{0.5}Cu_2$ doped with 5 at. % Gd is blurred. One of the reasons for this is possibly the spatial dispersion of the RKKY interaction. The resonant parameters, i.e, the g-factor and the spin relaxation rate of each individual Gd ion, are determined by averaging the contributions of all the Yb ions located in its immediate vicinity. When the concentration of Gd is increased from 2 to 5%, the deformation of the crystal lattice increases. The g-factor shift is determined by the first moment of the line shape, and ΔH is determined by the second moment; therefore Δg is more sensitive to variation of the mean distance between the deformed volumes than is ΔH. A more detailed RKKY theory, which takes into account the spatial dispersion and the influence of the mean distances between the Gd^{3+} and Yb^{3+} and between the Gd^{3+} and Gd^{3+} ions, is needed for quantitative evaluations.

The ESR linewidth ΔH reflects the temperature dependence of the product $k_B T\chi$, i.e., a linear behavior is observed when χ does not depend on the temperature, and a tendency to achieve saturation is observed when the static susceptibility follows Curie's law.

The values of $\Delta H(T)$ and $\Delta g(T)$ are usually described by two types of interactions: RKKY exchange and, when there is a linear dependence of $\Delta H(T)$, the Korringa relaxation mechanism. Let us consider both types of interactions. The spin of a Gd^{3+} ion interacts with conduction electrons by direct exchange and with Yb ions by means of RKKY coupling:

$$H = \sum_k J_0 \mathbf{S}\sigma_k + \sum_i \mathbf{S}\cdot\mathbf{S}_f \Gamma_{\text{Gd-Yb}}(\mathbf{R}_i), \tag{6}$$

where $\mathbf{S}$ is the spin of the Gd^{3+} ion, σ_k is the spin of the conduction electron, $\mathbf{S}_f$ is the spin of the Yb ion, J_0 is the exchange integral of Gd^{3+} ions with conduction electrons, $\Gamma_{\text{Gd-Yb}}(\mathbf{R}_i)$ is the effective RKKY exchange constant of Gd and Yb ions separated by a distance $\mathbf{R}_i$, and the summation is carried out over k conduction electrons and i Yb ions. In the free-electron model, the effective exchange constant equals

$$\Gamma_{\text{Gd-Yb}}(\mathbf{R}_i) = -3\pi z J_0 J_f N(E_F) F(2k_F R), \tag{7}$$

where z is the number of electrons in the atom, J_f is the exchange integral for the interaction of the Yb ions with the conduction electrons, $F(2k_F R) = \cos(2k_F R)/R^3$ is the oscillatory RKKY function, and $N(E_F)$ is the electronic density of states at the Fermi level. We denote this expression by

$$\Gamma = \sum_i \Gamma_{\text{Gd-Yb}}(\mathbf{R}_i) \sim \left(\sum_i \Gamma^2_{\text{Gd-Yb}}(\mathbf{R}_i) \right)^{1/2},$$

and this constant of the exchange integral will be determined experimentally.

When the spin relaxation rate is calculated, the first term leads to the familiar Korringa mechanism, while the second term leads to renormalized exchange of the paramagnetic Gd^{3+} and Yb^{3+} ions. Both mechanisms were considered by Coldea *et al.*.[7] in their ESR study of $Y_{1-x}Ce_xAl_2$ (see also Ref. 8). However, the expression derived by Coldea *et al.* for the RKKY relaxation rate must be modified slightly for our case. First, the splitting of the octet of the Yb^{3+} ion into a

lower Γ_6 doublet, an excited Γ_8 quartet, and an upper Γ_7 doublet by the cubic crystal field must be taken into account. Second, the interaction of the Gd ions with Yb ions should be represented as the product $\mathbf{J}\cdot\mathbf{S}$:

$$H_{\mathrm{RKKY}}=(g_f-1)\sum_i \mathbf{J}\cdot\mathbf{S}\Gamma_{\mathrm{Gd-Yb}}(\mathbf{R}_i). \tag{8}$$

Using the expressions of Coldea *et al.*, we obtain

$$\Delta g=J_0N(E_F)+\frac{g_f-1}{g_f}\frac{\chi}{\mu_B^2}\sum_i \Gamma_{\mathrm{Gd-Yb}}(\mathbf{R}_i), \tag{9}$$

$$\Delta H(T)=\Delta H_k+\frac{(g_f-1)^2}{g_f^2}\frac{2k_BT}{g\mu_B^3h}\chi\tau\sum_i \Gamma^2_{\mathrm{Gd-Yb}}(\mathbf{R}_i), \tag{10}$$

$$\Delta H_K=\frac{\pi(J_0N(E_F))^2k_BT}{g\mu_B}, \tag{11}$$

where τ is the spin relaxation time of the Yb ions and χ is the magnetic susceptibility of the Yb ions with consideration of x, i.e., their fraction in the lattice. The spin relaxation of the Yb ions describes the interaction of the fluctuating spin with the conduction electrons. The fluctuations occur with a high frequency and result in narrowing of the line.

The temperature dependences of the ESR linewidth and the g factor were calculated using Eqs. (9)–(11). However, it turned out that it is impossible to achieve agreement with experiment. Two difficulties arose. First, as we have already mentioned, the linear dependence of the linewidth is usually attributed to the Korringa mechanism. If this tradition is followed and $J_0N(E_F)$, which is sometimes called the Kondo coupling constant, is evaluated from the Korringa slope $\Delta H(T)/T=B=12.5$ G/K, we obtain 0.023. The value from the ESR shift is $\Delta g=0.045$, i.e., it is two times greater. Second, a more serious difficulty arises when an attempt is made to extrapolate the linear dependence of the linewidth to high temperatures. At high temperatures the extrapolated line intersects the experimental curve, i.e., the real relaxation rate is significantly smaller than the Korringa rate. Therefore, it must be assumed that the value of the Korringa exchange integral J_0 is small and the linewidth is determined completely by the RKKY interaction. It is possible that the Korringa mechanism is weak because of the low normal (unenhanced) density of states of the conduction electrons.

In fact, the measurements of the specific heat performed in Ref. 3 on $Yb_{0.4}In_{0.6}Cu_2$ and the isomorphous compound $Lu_{0.4}In_{0.6}Cu_2$ showed that the value of the density of states due to the light s band is 100 times smaller in $Lu_{0.4}In_{0.6}Cu_2$ than in $Yb_{0.4}In_{0.6}Cu_2$ where the density of states is determined by the f electrons. Hence, the normal density of states can be considered vanishingly small, and the Korringa contribution to the relaxation rate and to the g-factor can be neglected. As a result, only the second terms are left in Eqs. (9) and (10). Thus, the following expressions are used to describe the g-factor and the ESR linewidth

$$\Delta g=\frac{g_f-1}{g_f}\frac{\chi x\Gamma}{\mu_B^2}, \tag{12}$$

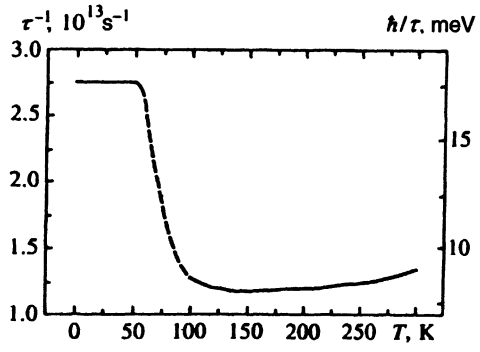

FIG. 5. Temperature dependence of the spin relaxation rate τ^{-1} of the Yb^{3+} ions in $Yb_{0.5}In_{0.5}Cu_2$ doped with 2 at. % Gd obtained from measurements the linewidth.

$$\Delta H=\frac{(g_f-1)^2}{g_f^2}\frac{2k_BT\chi x\Gamma}{g\mu_B^3h}\tau. \tag{13}$$

We stress, however, that the physical meaning of the magnetic susceptibility appearing in Eqs. (12) and (13) is significantly different.

Above the phase-transition point the Yb^{3+} ions form a system of almost independent localized magnetic moments. According to Sugawara,[13] the relaxation of the spins of dopant ions in such a material is determined by fluctuations associated only with the orientational part of the susceptibility (4), while the g factor is described by the total static susceptibility (3), including the Van Vleck part.

From the large values of $\chi(0)$ and $\gamma(0)$, it can be postulated that below the phase-transition point, i.e., in the ground state, the Yb^{3+} ions form a dense Kondo lattice. In this state the spins of the ions are completely shielded by the conduction electrons. The magnetic susceptibility of a dense Kondo state is temperature-independent and is specified by (2), and the same value of $\chi(0)$ appears in (12) and (13). It is noteworthy that Eq. (13) practically coincides with the results reported by Cox,[12] who calculated the relaxation rate of the magnetic moments of ions in a heavy-fermion material.

The formulas (12) and (13) are attractive, because they contain only two unknowns, which can be determined experimentally from the values of the ESR linewidth and the g-factor shift: Γ and τ. They were adjusted so as to obtain the best agreement with experiment.

The results of the calculations are represented by the solid lines in Figs. 2 and 3 for high and low temperatures. A reasonable value was obtained for the RKKY exchange integral: $\Gamma=2.9$ meV.

Figure 5 presents the temperature dependence of the spin relaxation rate. At high temperatures τ^{-1} is weakly dependent on the temperature. The value at low T, $\tau^{-1}=1\times10^{-13}-3\times10^{-13}$ s^{-1}, is of the order of the rate of the fluctuations found in other heavy-fermion materials. An evaluation of the Kondo temperature gives $T_K=\hbar/\tau\simeq18.1$ meV$=210$ K. As expected, this value is greater than the splitting by the cubic field ($\Delta=135$ K).

As we have already mentioned, Rajan[11] calculated the temperature dependence of the static magnetic susceptibility

for a Kondo system with an arbitrary spin J. For $J>1/2$ the plot of $\chi(T)$ has a maximum at $T_m=0.2T_K$. It is noteworthy that this temperature is close to the phase-transition point $T_m\sim T_p\sim 40$–50 K. This may not be a coincidence, and, perhaps, the increase in the susceptibility stimulates the phase transition.

Finally, let us examine the question of the nature of the first-order phase transition in $Yb_{0.5}In_{0.5}Cu_2$ Although the Felner–Nowik model, which is based on the variation of the valence of the Yb ions, provides a satisfactory description of many properties of this material, we believe that the approach based on the conception of a Kondo lattice in the low-temperature phase is preferable.

The following arguments can be brought to support our position:

1. The value of T_K which we obtained is considerably greater than the effective s–f hybridization temperature $T_f=5$ K in the Felner–Nowik model. This is inconsistent with the hypothesis that the phase transition is accompanied by a significant change in the valence of the ytterbium ions. In fact, the x-ray measurements in Ref. 3, showed that the phase transition is accompanied by only a slight change in valence (from 2.8 below the phase transition to 2.9 above it).

2. To interpret our ESR data, we postulated that $Yb_{0.5}In_{0.5}Cu_2$ is a heavy-fermion system. Evidence that the model which we have chosen is correct is provided by the good agreement between the calculations and experiment both below and above the phase-transition point. The linear dependence of $\Delta H(T)$ at low temperatures, which is caused by renormalized Korringa relaxation, attests to the existence of a Kondo state in this temperature range. It should be stressed that both of the constants which we obtained, i.e., the RKKY exchange integral J_{Gd-Yb} and the spin relaxation rate τ^{-1}, have perfectly satisfactory numerical values.

3. The appreciable increase in the relaxation rate τ^{-1} below the phase-transition point indicates an increase in the Kondo correlations.

The mechanism of the phase transition in mixed-valence and heavy-fermion systems was discussed by Chandran *et al.*[14] There are two possibilities: a dependence of the volume on the position of the f level (measured from the Fermi energy) and a dependence of the volume on the Kondo coupling constant, i.e., on the electronic density of states (in this case the positive feedback is observed owing to the nonlinear dependence of the Kondo temperature and the Kondo energy on the coupling constant).

It was shown that the first possibility leads to a mixed-valence state and a phase transition with a large change in valence, i.e., to the Felner–Nowik model. The second possibility leads to Kondo volume collapse similar to the phase transition in Ce previously considered by Allen and Martin. We believe that the phase transition in $Yb_{0.5}In_{0.5}Cu_2$ takes place by just this mechanism.

4. CONCLUSIONS

The static magnetic susceptibility, the ESR linewidth, and the g-factor of Gd^{3+} ions in the intermetallic compound $Yb_{0.5}In_{0.5}Cu_2$ have been measured both below and above the phase-transition point. The relaxation rate is determined mainly by the interaction between the Gd and Yb ions.

Our results attest to the many-particle nature of the ground state of $Yb_{0.5}In_{0.5}Cu_2$ at low temperatures. It exists in the form of a dense Kondo lattice of Yb^{3+} ions, which causes renormalized Korringa relaxation. At high temperatures the system passes into a state with well defined localized magnetic moments for the Yb^{3+} ions and is described by an RKKY interaction. It may therefore be postulated that the phase transition in $Yb_{0.5}In_{0.5}Cu_2$ has much in common with the well known α–γ transition in metallic cerium and takes place owing to the Kondo volume collapse predicted by Allen and Martin.

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