made *in situ* on the sample in the ESR cavity. We observe the separation between the ESR signals from a pair of thin DPPH spin markers placed 90° apart on the central plane of a cylindrical sample. The details of the technique, computer simulations, experimental checks via a Faraday susceptometer, etc., will be presented in a future publication. The samples were prepared in an arc furnace, annealed under argon at 1100 K for 24 h, spark cut into cylinders, and heavily etched. Both quenched and slowly cooled samples produced consistent results.

[2]P. Monod and Y. Berthier, J. Magn. Magn. Mater. 15–18, 149 (1980).

[3]J. Owen, M. E. Brown, V. Arp, and A. F. Kip, J. Phys. Chem. Solids 2, 85 (1957).

[4]We associate only one direction with the order parameter for the sake of simplicity. We omit gradient terms because we are concerned only with the uniform response (wave vector = 0).

[5]I. E. Dzyaloshinski and G. E. Volovick, Ann. Phys. (N.Y.) 125, 67 (1980).

[6]The two modes correspond to the two circular polarizations of $\delta\vec{M}$, with (+) corresponding to the customary polarization sense in magnetic resonance. $\omega^{-}>0$ for H such that $\hat{n}=\hat{N}$ is the stable equilibrium.

[7]Kichi Okuda and Mineyuki Date, J. Phys. Soc. Jpn. 27, 839 (1969).

[8]Should more accurate measurements confirm that the magnitude of the slope is less than $\frac{1}{2}$, it would imply that $\chi_{\parallel}>\chi_{\perp}$ in this alloy.

[9]Equation 2 can be used to derive $\chi'(\omega)$, the real part of the rf susceptibility for linearly polarized rf magnetic field. Recall that the dc field H is varied; the singular part of $\chi'(\omega)$ turns out to be $(\chi\omega/2r)(H-H_r)^{-1}$ for $\omega/\gamma>H_i$, and $(\chi\omega/2r)(H_r-H)^{-1}$ for $\omega/\gamma<H_i$, where H_r is the field for resonance: $\gamma H_r=|\omega^2-(\gamma H_i)^2|/\omega$.

[10]Note that when measured in fields $\simeq 3$ kG, $T_g'\simeq 0.9T_g$ (T_g determined at low fields is given by $9.5\,C_{\mathrm{Mn}}{}^{0.65}$). We use T_g' in our relation because experimentally it is also the temperature where the ESR anisotropy effects (as measured in fields = 3 kG) have gone to zero. We also find that with the addition of 0.3 at.% Ni to CuMn alloys there is no change in T_g' to within ± 5%.

[11]Values of K determined from hysteresis-loop areas agree with our scaling but are smaller as would be expected. P. Monod, J. Prejean, and B. Tissier, J. Appl. Phys. 50, 7324 (1979). Values of K deduced from zero-field NMR agree with our values. H. Alloul, J. Appl. Phys. 50, 7330 (1979).

[12]W. M. Saslow, Phys. Rev. B 22, 1174 (1980).

ESR Study of the Kondo Effect in *Au*:Yb

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(Received 4 June 1980)

Paramagnetic resonance experiments at 3 and 9 GHz were performed in dilute *Au*:Yb in the temperature range from 100 mK to 4 K. All results show for the first time a logarithmic temperature dependence for the g-value shift and for the relaxation rate in complete agreement with theoretical predictions. The fit yields a Kondo temperatue of $T_K\sim 10$ μK and a degeneracy of $d\sim 3$ for the local-moment–conduction-electron interaction channels.

PACS numbers: 75.20.Hr, 76.30.Kg

It has been pointed out by Orbach[1] that the ESR of local moments could, in principle, be used to observe the Kondo effect directly. The change of the local susceptibility and the relaxation rate of local moment could be determined independently in one experiment. However, the ESR will be limited to a temperature range $T>T_K$, where the local moment is well defined. Most of the experiments in the field of Kondo effect in dilute alloys did measure the static properties (susceptibility, thermopower, etc.) or the scattering rate (T-matrix) via the resistivity anomaly. Only a few experiments determined the relaxation rate of the local moment, e.g., neutron scattering[2] and NMR[3] in *Cu*:Fe or Mössbauer effect[4] in *Au*:Yb.

The first observation of the ESR for the *Au*:Yb system was made by Hirst *et al.*,[5] where the g-value shift was shown to be negative, indicating that the interaction between the conduction electrons of the host and the Yb impurities is antiferromagnetic. In this Letter we present the first experimental evidence of the Kondo effect in the linewidth and the g shift in an ESR spectrum. We choose *Au*:Yb because the ESR of this system is not bottlenecked and the full effect on the exchange should be visible.

The commonly accepted description for a nonbottlenecked ESR is given by[6]: (1) g shift, $\Delta g=g_{\mathrm{metal}}-g_{\mathrm{ionic}}=\alpha N(E_F)J_1$, and (2) relaxation rate of the local moment to the conduction electrons, $\hbar\tau^{-1}=\hbar\delta_{ie}=\pi\alpha^2[N(E_F)J_2]^2kT$, where α is the modified de Genne factor $\alpha=(g_J-1)g_J{}^{-1}g_{\mathrm{eff}}=0.418$

(for *Au*:Yb). At this point we assume an isotropic exchange $\mathcal{H}_{ex} = -J\vec{S}\cdot\vec{\sigma}$. The exchange parameters J_1 and J_2 usually do not agree. This can be caused by many reasons outlined in Ref. 1. We will show that the present experiment sheds some light into this problem, especially the degeneracy of the relaxation path.[7] For $T > T_K$ the most transparent way to include higher-order correction into the exchange coupling is to treat the effective coupling strength as being temperature dependent[8]: $N(E_F)J_{eff}(T) = [\ln(T_K/T)]^{-1}$, $T_K = D\exp[1/N(E_F)J_0]$. This is only correct for one "interaction channel"; and for $2l+1 = d$ channels, T_K transforms to $T_K = D\exp[d/N(E_F)J_0]$ and $N(E_F)/d$ can be seen as a partial density of states N_ν. The corresponding formulae for ESR are

$$\Delta g = \alpha \sum_{\nu=1}^{d} N_\nu J_1(T) = \alpha d/\ln(T_K/T), \quad (1)$$

$$\hbar\delta_{ie}/\pi kT = \alpha^2 \sum_{\nu=1}^{d} [N_\nu J_2(T)]^2 = \alpha^2 d[\ln(T_K/T)]^{-2}. \quad (2)$$

For simplicity we have assumed an incoherent summation over the d channels. Hence the only parameters to be determined by the experiment are T_K and d. T_K for *Au*:Yb has already been determined to be much smaller than 10 mK.[4,9] Because of the low T_K, experiments in the millikelvin regime and over a wide range in temperature (factor of 40) were needed. On the other hand, finite magnetic fields reduce the temperature dependence. In a rough estimate, T can be replaced by $(T^2+H^2)^{1/2}$. Subsequently the conventional microwave of 9 GHz ($\cong$430 mK) is not as useful as 3 GHz ($\cong$140 mK).

Our experiments were performed using a combination of a ^{3}He/^{4}He dilution refrigerator and commercial microwave units, and only the sample was cooled down.[10] Special care was taken for sample preparation since most of the previous ESR experiments on *Au*:Yb show[11,12] a rather large linewidth $\Delta H = a + bT$, $bT = \hbar\delta_{ie}(g_{eff}\mu_B)^{-1}$, with a residual linewidth a of the order of 100 G. This deforms the line shape and makes a precise determination of H_{res} and ΔH impossible. A master alloy of 0.5 at.% Yb in Au was first prepared from which the less concentrated alloys were made by diluting the master alloy with Au. We grew single-crystal and polycrystalline rods (5 mm in diameter and 2 cm long) by the Bridgman method. This technique reduces the residual linewidth as compared to arc-melted foils, etc.,[10,13] by a factor of 2 or 3 (see Table I). [Before induction melting of the buttons, the samples were cleaned by etching with a solution of HCl-HNO_3, and the same etching procedure was used before each ESR measurement. The concentration of the samples was determined by measuring the saturation magnetization to 50 kG and 1.5 K, taking into consideration the excited crystal-electric-field levels.]

Figure 1 shows an experimental spectrum. The solid line is the theoretical Dysonian line shape with $A/B = 2.53$. The perfect agreement between theoretical shape and experiment, and the narrow linewidth enables us to determine small changes in H_{res} and ΔH. No enriched ^{170}Yb isotope was needed because the hyperfine lines of isotopes 171 and 173 appear only in the wings and have been put into the theoretical fit. The solid lines

TABLE I. Results of fitting experimental data with Eqs. (1) and (2).

Method	d	T_K (μK)
all g-value shifts	3.8 ± 1.2	0.5 – 6
linewidth, 3.22 GHz		
500 ppm, a = 15.0 G;		
1200 ppm, a = 22.5 G;		
2400 ppm, a = 50.0 G	2.4 ± 0.8	1 – 10
linewidth, 8.92 GHz		
500 ppm, a = 32.0 G;		
1200 ppm, a = 39.0 G	2.5 ± 0.8	3 – 14
linewidth, 8.92 GHz		
2400 ppm, a = 95.0 G	2.8 ± 0.8	0.7 – 10

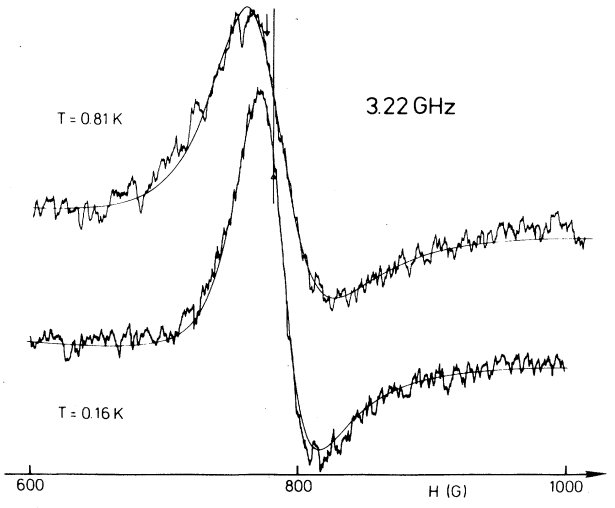

FIG. 1. Spectra of 1200-ppm Yb in Au. The arrows indicate the shift of the resonance field as a function of temperature. $\tau = 0.3$ sec is used.

are computer simulation from which accurate values for H_{res} and ΔH can be extracted. The experimental g values are shown in Fig. 2 where, within a certain scatter, all data points show a common trend that is independent of concentration and frequency. The g value at 4 K agrees well with previously published data and then it drops slowly from 4 to 1 K. Below 0.7 K there is a rather sharp drop in g value. A plot of $(\Delta g)^{-1}$ vs $\ln T$ yields $\ln T_K$ as the intercept and d from the slope with a degree of fit better than 0.97.

$$\hbar\delta_{ie} = \pi\alpha^2[N(E_F)J_2]^2[\tfrac{1}{2}kT(\langle+|J_z|+\rangle - \langle-|J_z|-\rangle)^2 - \tfrac{1}{4}\hbar\omega\coth(\hbar\omega/2kT)(\langle+|J_+|-\rangle)^2]. \quad (3)$$

A plot of $[(\Delta H - a)/T]^{-1/2}$ vs $\ln T$ [see Eq. (2) and text] gives again a straight line for high temperatures (Fig. 3, full line) and deviation from linearity is corrected by using Eq. (3) (dashed line). From the figure one sees that at high Yb concentrations (solid triangle, open circle), spin-spin interaction occurs below 0.7 and 0.3 K, respectively, which increases the linewidth and causes departure from the fit. For the 500-ppm sample (open square) agreement with the fit (dashed line) is excellent; it is even better for the 9-GHz results over the whole temperature range of 1.01 to 4 K.

We would like to stress again that up to this point the analysis of the data fits T_K and d independently from linewidth and g-value shift. No separate choice of the parameter D, the density of states $N(E_F)$, and J_1 and J_2, was necessary.

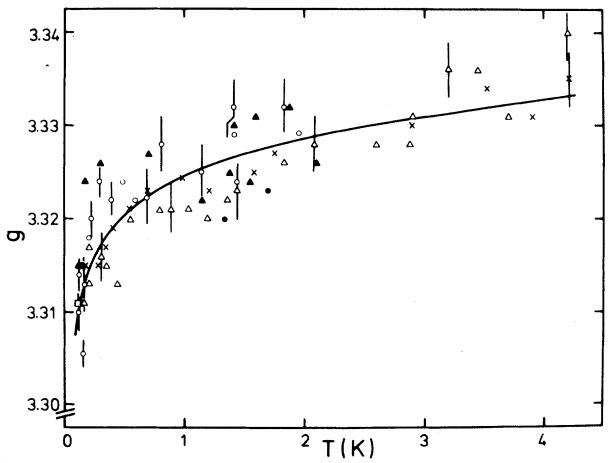

FIG. 2. Plot of experimental g value vs temperature. Open triangles, 2400 ppm, 9 GHz; solid triangles, 2400 ppm, 3 GHz; solid circles, 1200 ppm, 9 GHz; open circles, 1200 ppm, 3 GHz; crosses, 500 ppm, 9 GHz; and square, 500 ppm, 3 GHz. Solid line is generated by taking $d = 3.8$ and $T_K = 4$ μK.

At resonance field of only 700 G the admixture of excited levels into the Γ_7 ground state can be neglected.

The analysis of the linewidth is more complicated: (1) The residual linewidth a is in principle an unknown fitting parameter; (2) at 1200 and 2400 ppm, and low temperatures, Yb-Yb interaction will influence ΔH also; (3) since we are not in the high-temperature limit ($\hbar\omega \approx kT$) even in first order, a deviation of linear T dependence appears[14]

Using the naive picture of an isotropic exchange, equal weighting of the interaction channels, and the incoherent sum over $(N_\nu J)^2$ one would not expect T_K and d to be the same from linewidth and g-value shift (Table I). The range in T_K is due to extrapolation over four decades in temperature, the large error bar of d to fitting all g value of six experiments together. All experiments together yield a Kondo temperature $T_K \sim 10^{-5}$ K and a degeneracy factor of $d \sim 3$. Previous ESR measurements, to the best of our knowledge, did only combine g-value shift and Korringa rate and determine $(J_2/J_1)^2 = d$. This can be quite hazardous as pointed out earlier since many factors cause $J_2 \neq J_1$. The Mössbauer relaxation data[4] are within the error bars of both experiments and in rough agreement with our results: Both show an increase of the relaxation/temperature of $\cong 70\%$ between 4 and 200 mK. However, the analysis in Ref. 4 is slightly different from Eq. (2).

The cutoff parameter D is difficult to choose since it is not a physical observable. Values between the Fermi energy and a "narrow" d-band width are commonly chosen. If we do so, one gets for $T_K = 10^{-5}$ K, $N_\nu J_0(D = 10^4\ \text{K}) = -0.047$, and $N_\nu J_0(D = 10^2\ \text{K}) = -0.06$, respectively, whereas the effective exchange in the g-value shift at 1 K $N_\nu J_{eff}(T = 1\ \text{K}) = -0.09$. This difference of calculated bare exchange and the experimental effective one has already been pointed out for systems with ferromagnetic coupling.[15]

In summary we employ low concentration, low temperature, and low-microwave ESR to keep the experimental line shape as narrow as possible and perfect Dysonian-like.[16] This enables us to show the logarithmic temperature dependence in g-value shift and linewidth. Even with the limitation of a much simpler theory based on isotropic exchange interaction and an isolated Γ_7 state,

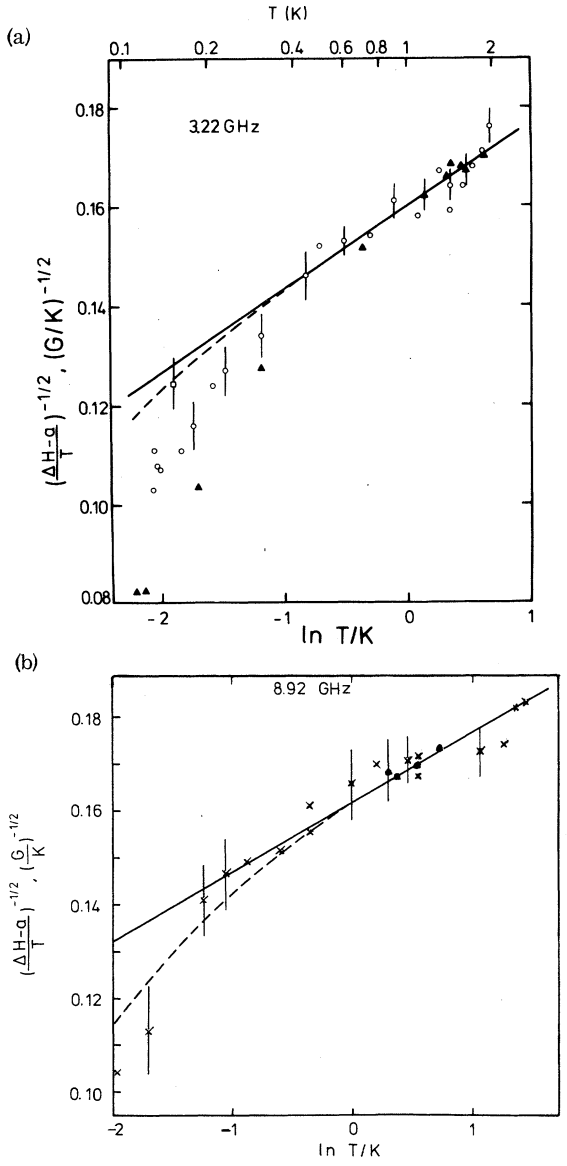

FIG. 3. (a) A fit of experimental linewidth at 3 GHz to Eq. (2). Solid line gives fit and dashed line gives that corrected for relaxation because of Eq. (3). (b) Same fit as Fig. 3(a) for 9 GHz, 1200 and 500 ppm. The applied field of 1.9 kG reduces the relaxation by a few percent (Ref. 17). This could not have been observed in our experiment with the given error bars; it should have ***reduced*** the slope of the solid line.

good agreement between data and theory is obtained in the present investigation.

The authors are indebted to Dr. S. E. Barnes for theoretical help and continuous interest, and for helping us through the "Kondo wilderness". Y. v. Spalden contributed to experimental work and the authors also wish to acknowledge discussions with Professor L. L. Hirst, Professor R. Orbach, Professor D. Riegel, Professor R. Schlottman, and Professor K. D. Schotte. This work was supported by the Deutsche Forschungsgemeinschaft special research fund (Sondersforschungsbereich 161).

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[1]R. Orbach, in *Low Temperature Physics, LT-14*, edited by M. Krusius and M. Vuorio (American Elsevier, New York, 1975), Vol. 5, p. 375, and references therein.

[2]M. Loewenhaupt and W. Just, Phys. Lett. 53A, 305 (1975).

[3]H. Alloul, Physica 86–88B, 449 (1977); O. Kanert, M. Mali, K. Preusser, and M. Mehring, Solid State Commun. 21, 1047 (1977).

[4]F. Gonzales-Jimenez and P. Imbert, Solid State Commun. 13, 85 (1973).

[5]L. L. Hirst, G. Williams, D. Griffiths, and B. R. Coles, J. Appl. Phys. 39, 844 (1968).

[6]For notations and explanations of symbols, see Ref. 1.

[7]B. Caroli, J. Phys. F 5, 1399 (1975).

[8]P. W. Anderson, Comments Solid State Phys. 1, 31 (1968), 1, 190 (1968); S. E. Barnes, J. Phys. F 6, 1713 (1976).

[9]A. Benoit, J. Flouquet, and J. Sanchez, Phys. Rev. B 9, 1092 (1974).

[10]J. Nagel, K. Baberschke, and E. Tsang, J. Magn. Magn. Mater. 15–18, 730 (1980).

[11]L. J. Tao, D. Davidov, R. Orbach, and E. P. Chock, Phys. Rev. B 4, 5 (1971).

[12]J. Nagel, S. Hüfner, and M. Grünig, Solid State Commun. 13, 1297 (1973).

[13]Y. v. Spalden, Diplom thesis, Freie Universität, Berlin, 1979 (unpublished), and to be published.

[14]L. L. Hirst, Phys. Rev. 181, 597 (1969).

[15]B. Bachor, K. Baberschke, and S. E. Barnes, Phys. Rev. B 21, 2666 (1980).

[16]The analysis of the residual linewidth is not an issue of the present Letter, but agrees well with $a=\alpha c+\beta\nu+\delta c\nu$, and is the subject of Ref. 13.

[17]A. K. Bhattacharjee and B. Coqblin, Solid State Commun. 18, 1587 (1976).