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Hyperfine Splitting of Er and Yb Resonances in Au: A Separation between the Atomic and Covalent Contributions to the Exchange Integral*

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The hyperfine splitting has been observed in the magnetic resonance spectrum of dilute Au:Er and Au:Yb alloys. In both cases, the observed hyperfine coupling constant was larger (more positive) than in cubic nonmetallic hosts. Attributing this increase to conduction-electron polarization, and utilizing independent estimates of the 6s contribution to the hyperfine coupling constant, the atomic and covalent contributions to the localized-conduction-electron exchange integral are separated. It is found that the latter increases by an order of magnitude from Er to Yb in Au. This increase is attributed to the nearness of the $4f^{14}$ level to the Fermi surface in Au:Yb alloys, noting that Ag:Yb is diamagnetic.

I. INTRODUCTION

We report the first observation of the hyperfine splitting in the magnetic resonance spectrum of dilute Au:Yb and Au:Er alloys. In both cases the hyperfine coupling constant A is larger (more positive) than in nonmetallic hosts with the same local symmetry. This increase is attributed to the spin-polarized conduction electrons. The temperature dependence of the resonance linewidth yields values for the localized-conduction-electron exchange coupling constant J . Knowing the increase in A and J over the insulator value, and using the estimate of Gossard *et al.*¹ of the value of the 6s contribution to the hyperfine coupling constant, we are able to separate out the two contributions to J , commonly referred to as "atomic" (J_{at}) and "covalent mixing"

(J_{cm}), for both alloys. We find the ratio of (J_{cm})_{Au:Yb} to (J_{cm})_{Au:Er} to be approximately 20, much larger than expected from simple theoretical estimates.² We speculate, knowing that the isomorphous alloy Ag:Yb is diamagnetic,³ that this is caused by a close proximity of the $4f^{14}$ virtual level in Au:Yb to the Fermi level, allowing a strong "absorption" contribution² to J_{cm} to obtain for that alloy.

II. EXPERIMENTAL RESULTS

A. Au:Er

The magnetic resonance spectra of powdered samples of Au:Er for Er concentrations of 50, 100, 500, 1000, and 5000 ppm was obtained as a function of temperature. The measurements were conducted at 3-cm wavelength, the apparatus having been described elsewhere.⁴ The spectrum of a 50-ppm

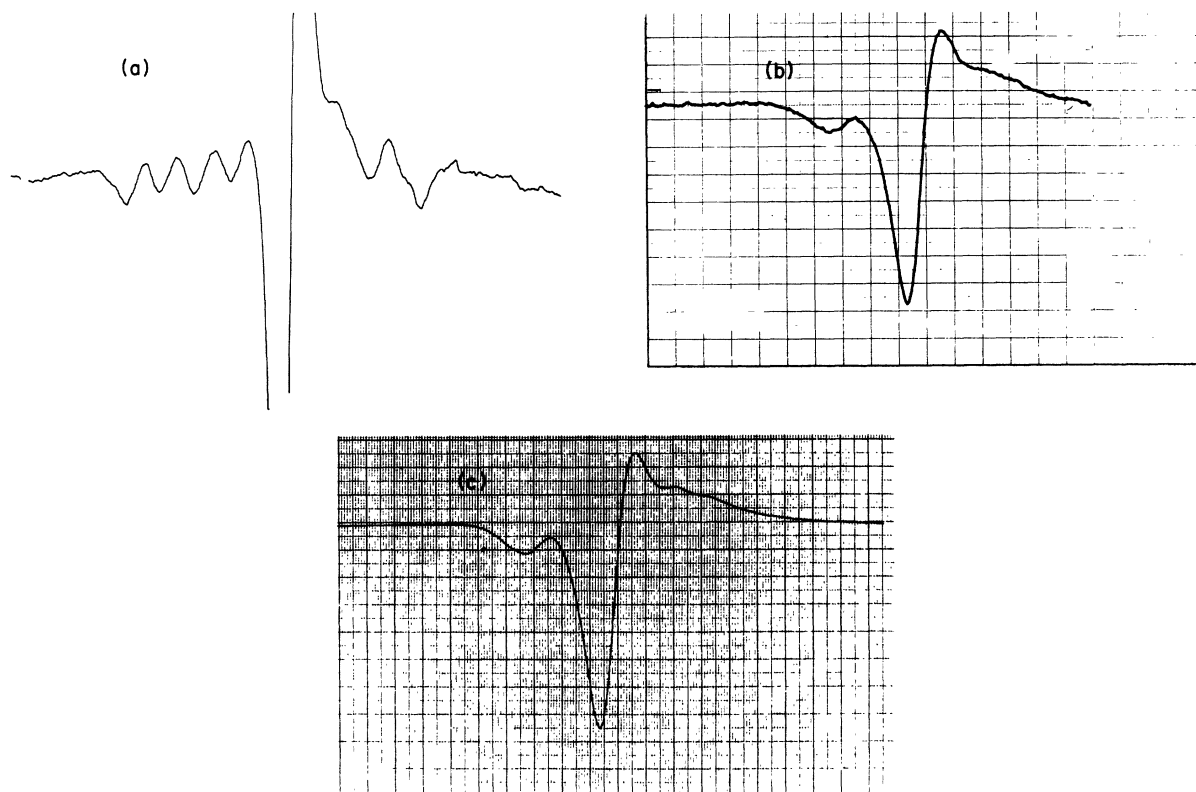


FIG. 1. (a) Electron spin resonance spectrum of a powered sample of 50 ppm of Er in Au at 1.4 °K at X band. The central line corresponds to Er isotopes with $I=0$. The surrounding satellites (seven of them are clearly seen, the eighth is hidden by the central line) arise from the Er^{167} isotope with $I=\frac{7}{2}$. The spectrum can be fitted by a hyperfine coupling constant $A=75.5\pm 0.5$ G. (b) Resonance spectrum of a powdered sample of 500 ppm of Yb in Au at 1.4 °K at X band. The central line corresponds to Yb isotopes with $I=0$. The two satellite lines arise from the Yb^{171} isotope with $I=\frac{1}{2}$. The intensity of the Yb^{173} isotope (with $I=\frac{5}{2}$) lines is too small to be observable. (c) A theoretical spin resonance spectrum for Yb in Au. In deriving this synthesis, we have included all the hyperfine satellites (corresponding to $I=0, \frac{1}{2}, \text{ and } \frac{5}{2}$) with intensities appropriate to their natural abundances, and with a common residual and thermal width. Each line is a sum of the form $\chi'(X_i) + \alpha\chi''(X_i)$, where $\chi''(X_i)$ is the absorption derivative and $\chi'(X_i)$ is the dispersion derivative appropriate to a Lorentzian line. The quantity X_i is defined by $X_i = (H - H_{\text{res}})/\Delta H$, where ΔH is the measured linewidth at 1.4 °K. A best fit obtains for $\alpha=1$, appropriate to an A/B ratio of 2.7. The value of H_{res} for each of the Yb^{171} hyperfine lines was changed continuously to fit the observed spectrum (with appropriate positions and width) and a best fit was obtained for $A^{171}=575\pm 10$ G.

sample at 1.4 °K is exhibited in Fig. 1(a). This spectrum exhibits the same character as that of Ag:Er published previously.⁵ A strong "central" line (corresponding to Er isotopes with $I=0$) is surrounded by at least six hyperfine line satellites (corresponding to the Er^{167} isotope, with $I=\frac{7}{2}$). The ratio of combined intensities of the hyperfine satellites to the central line agrees with the natural abundance of Er^{167} (24%). The form of the central line exhibits the metallic resonance shape with an amplitude-peak ratio⁶ approximately equal to 2.0.

Using the second-order hyperfine splitting formula,

$$H = H_0 - AM_I - (A^2/2H_0)[I(I+1) - M_I^2],$$

as well as a superposition line-shape analysis, we have estimated the resonance line positions. The fitted values are appropriate to a hyperfine constant $A=75.5\pm 0.5$ G. The reported hyperfine constants for Er in various insulators⁷⁻⁹ are listed in Table I. These results indicate that the hyperfine constant in metals is consistently larger (more positive) than in insulators.

The resonance field of the central ($I=0$) line corresponds to a g factor of 6.80 ± 0.04 . The linewidth of the main resonance is of the form $a(c, H) + bT$, where $b=2.4\pm 0.2$ G/deg and a , the residual linewidth, increases with concentration c and magnetic field H . A detailed examination of the residual linewidth will be given elsewhere.

TABLE I. Comparison between the hyperfine constants and the g values for Er and Yb in various cubic metals and insulators.

Host	$g(I=0)$ expt	$A(G)$ expt	$\Delta A(G)$ expt	$J(eV)$	Δg calc	g calc
Cubic insulators						
ThO ₂ :Er ^a	6.752 ± 0.005	73.8 ± 0.1				6.772
CeO ₂ :Er ^a	6.747 ± 0.01	73.4 ± 0.1				6.772
CdF ₂ :Er ^a	6.758	73.0 ± 0.1				6.722
CaF ₂ :Er ^a	6.785	71.2 ± 0.1				6.772
Cubic metals						
Ag:Er	6.84 ± 0.05	76 ± 1	~ 3	+0.16 ± 0.03 ^b	+0.06	6.83
Au:Er	6.80 ± 0.04	75.5 ± 0.5	~ 2.5	+0.1 ± 0.02	+0.02	6.79
Cubic insulators						
ThO ₂ :Yb ^a	3.423	548.8 ($I=\frac{1}{2}$)				3.423
CeO ₂ :Yb ^a	3.424	548.8 ($I=\frac{1}{2}$)				3.423
CaF ₂ :Yb ^a	3.438	549.2 ($I=\frac{1}{2}$)				3.423
Cubic metal						
Au:Yb	3.34 ± 0.06	575 ± 10 ($I=\frac{1}{2}$)	~ 26	-0.85 ± 0.2	-0.06	3.36

^aSee Refs. 7-12.^bIn calculating J for Ag: Er alloys, we use the value $b = 8$ G/deg and take into consideration exchange enhancement of the host metal.

B. Au:Yb

The magnetic resonance of Au:Yb has been observed for Yb concentrations of 500, 2000, and 5000 ppm.¹⁰ Alloys with lower concentrations exhibit very poor signals because of the relatively high residual ($T=0$) linewidth in these alloys.

Figure 1(b) exhibits the spectrum obtained for a 500-ppm sample of Yb in Au at 1.4 °K. A central resonance line (corresponding to Yb isotopes with $I=0$) surrounded by two hyperfine satellite lines (corresponding to the Yb¹⁷¹ isotope with $I=\frac{1}{2}$) is clearly seen. Using a superposition line-shape analysis, the central and hyperfine positions can be fitted with a hyperfine coupling constant $A = 575 \pm 10$ G and a g factor $g = 3.34 \pm 0.06$. This value is close to that expected for the Γ_7 ground state of Yb³⁺ in a cubic crystalline field. The linewidth of the main resonance is also of the form $a'(c, H) + bT$, where $b = 42 \pm 7$ G/deg and the residual width increases with concentration c and magnetic field H . The resonance of Yb³⁺ in various cubic nonmetallic hosts has been reported by many authors.^{7,11,12} Their results for A (listed in Table I) again are consistently smaller (less positive) than the value we report for the alloy. We did not observe in the metal resonance the spectrum appropriate to the Yb¹⁷³ isotope ($I=\frac{5}{2}$) which, however, was observed in the insulators. The ratio of the natural abundance of the isotopes is Yb¹⁷¹ ($I=\frac{1}{2}$): Yb¹⁷³ ($I=\frac{5}{2}$): Yb ($I=0$) = 14:16:70. The absence of the $I=\frac{5}{2}$ hyperfine lines in our measurements on Au:Yb alloys can be understood in terms of the small natural abundance of this isotope being

divided amongst six lines, together with the large residual width. Evidence for this conclusion is contained in Fig. 1(c), where a theoretical composite spectrum is synthesized. The g factor, linewidth, and hyperfine constant reported above, along with the line-shape ratio ($A/B=2.7$ here), were derived from a best fit of this synthesis to the experimentally observed resonance lines.

III. ANALYSIS

We assume that the orbital contribution to the g value of the paramagnetic ions in metals is the same as for nonmetallic systems (i. e., for f^{11} and f^{13} configurations g is uniquely determined by the Γ_7 component of the crystalline field split ground multiplet state). Therefore, any change of g in the metal relative to that of the insulator can be attributed to an exchange interaction of the type $J(\vec{S} \cdot \vec{s})$ with the conduction electrons, where \vec{S} is the true spin of the localized moments (in the Russell-Saunders sense) and \vec{s} the spin of a conduction electron. Such an interaction produces a Knight shift in the absence of a magnetic resonance bottleneck¹³ equal to

$$\Delta g = g [(g_J - 1)g_J] J\eta(E_F) . \quad (1)$$

In addition, the thermal broadening of the resonance line is given by¹⁴

$$b = \frac{\partial \Delta H}{\partial T} = \frac{\pi k g}{\mu_e} \left(\frac{g_J - 1}{g_J} \right)^2 [J\eta(E_F)]^2 , \quad (2)$$

where g_J is the Landé g factor, $\eta(E_F)$ the density of states per one spin direction per atom of the con-

duction electrons [for Au, $^{15} \eta(E_F) = 0.15 \text{ eV}^{-1} \text{ spin}^{-1} \text{ atom}^{-1}$] and k is the Boltzmann factor.

Substitution of the measured values of g , $\eta(E_F)$, and b into (1) and (2) enables us to obtain a value for the exchange coupling constant. The values of J so derived for the Au:Er and Au:Yb alloys are listed in Table I. The magnitude and sign of the exchange coupling constant can be understood in terms of a sum of a positive atomic exchange coupling J_{at} and a negative (covalent mixing) effective exchange coupling J_{cm} . The degree of mixing which determines the magnitude of the latter is strongly dependent on the $4f$ shell occupation number, and the proximity of the $4f^n$ and $4f^{n+1}$ energy levels to the Fermi energy. We shall assume J_{cm} is substantially larger for Au:Yb than for Au:Er. The atomic exchange is assumed to have the same value for both alloys because the Fermi energy remains the same.² It will turn out that, with these assumptions, the magnitude of the hyperfine coupling will allow us to estimate values for J_{at} and J_{cm} individually.

It has been stressed previously¹⁶ that when the states from which the doublet ground states are derived are eigenstates of the total angular momentum, the ratio of the orbital contribution to both A and g should be a constant, dependent only on the nature of the ground state. Therefore, we are led to the conclusion that the hyperfine constant A in metals should be the same as in nonmetals, except for additional contributions arising from the spin-polarized conduction electrons. The difference between A as measured in the alloys, and the cubic insulator, is termed ΔA and assumed to arise from two sources.

The exchange interaction polarizes the conduction band spins. This polarization affects the nuclear spin I of the paramagnetic ion by virtue of the contact hyperfine interaction $A_{\text{ce}}^{\text{contact}} (\vec{I} \cdot \vec{s})$. However, only the polarization arising from the atomic part of the exchange interaction, J_{at} , can contribute to the hyperfine constant via this term.¹⁷ The polarization produced by the covalent mixing interaction, J_{cm} , affects primarily only those electrons which are of f -like character (viewed from the impurity site)¹⁸ and therefore cannot contribute directly to a contact hyperfine field. However, a contribution can obtain via core polarization of the occupied inner atomic s shells. Therefore, the conduction-electron contribution to the hyperfine constant will take the form^{1,19,20}

$$\Delta A = \left(J_{\text{at}} \frac{A_{\text{ce}}^{\text{contact}}}{g_I \mu_I} + J_{\text{cm}} \frac{A_{\text{ce}}^{\text{cp}}}{g_I \mu_I} \right) \frac{g_I \mu_I}{g_e \mu_e} \frac{\eta(E_F)}{g}, \quad (3)$$

where $A_{\text{ce}}^{\text{contact}}/g_I \mu_I$ and $A_{\text{ce}}^{\text{cp}}/g_I \mu_I$ are the hyperfine field per spin for the contact and core-polarization terms, respectively. Gossard *et al.* assert¹ that

in metallic Yb the hyperfine field per spin is $A_{\text{ce}}^{\text{contact}}/g_I \mu_I = 6 \times 10^6 \text{ G}$ for the $6s$ electrons and $A_{\text{ce}}^{\text{cp}}/g_I \mu_I = -2.4 \times 10^6 \text{ G}$ for the core-polarization contribution arising from d electrons. We shall take over the *former* result for the alloys Au:Er and Au:Yb. Our argument is as follows. The orthogonalization theorem of Cohen and Heine²¹ suggests that, in the vicinity of the atomic core, the conduction electrons will assume a radial dependence not unlike the atomic valence electrons. In the case of rare-earth ions, this implies a $6s$ character to the s -like part of the conduction electrons in the immediate vicinity of the rare-earth core. Thus, for Au:Yb alloys, we use the value of Gossard *et al.*¹ for $A_{\text{ce}}^{\text{contact}}/g_I \mu_I$ which they derive from measurements on Yb metal. The insensitivity of this quantity to the $4f$ occupation number for the *same* effective screened nuclear charge allows us to use this value for Au:Er alloys as well. We recognize that this extrapolation may diminish the accuracy of our final results. We do not feel we are making gross errors, however, because experience with many metals²² gives one some confidence in such a procedure. Both J_{at} and $A_{\text{ce}}^{\text{contact}}$ are positive, while both J_{cm} and $A_{\text{ce}}^{\text{cp}}$ are negative, so that ΔA in (3) should always be positive, regardless of the relative magnitude of the two terms in the first parentheses, in agreement with a number of experimental results in other materials.¹⁹

For Au:Er we find ΔA to be approximately 2.5 G. The total exchange interaction J is positive, implying that J_{at} dominates over J_{cm} in this alloy. The value of $A_{\text{ce}}^{\text{contact}}$ is expected to be greater than $A_{\text{ce}}^{\text{cp}}$ so that, from (3), the combination of these two factors can allow us to neglect the second term in (3) as a first approximation. Substituting the measured value of ΔA and the value of Gossard *et al.*¹ of $A_{\text{ce}}^{\text{contact}}/g_I \mu_I$ into (3), we find $J_{\text{at}} = +0.15 \text{ eV}$. From Table I, the total exchange coupling constant determined from (1) and (2) equals $+0.10 \pm 0.02 \text{ eV}$, implying $J_{\text{cm}} = -0.05 \pm 0.02 \text{ eV}$. The latter value is very approximate because of the errors introduced when taking the difference of imprecisely determined large numbers.

Adopting the value of J_{at} derived above for Au:Yb alloys, and using the total exchange coupling constant of -0.85 ± 0.2 as listed in Table I, we find $J_{\text{cm}} = -1.0 \pm 0.2 \text{ eV}$. This very much larger value for J_{cm} for Yb as compared to Er in Au disagrees with previous theoretical estimates.² It can be understood in terms of the close proximity of the $4f^{14}$ virtual level to the Fermi level.³ This causes ΔE_{abs} [see Eq. (10a) of Ref. 2] to be small and leads, therefore, to a very large value for J_{cm} . The fact that Yb in Ag, as well as Yb metal, is diamagnetic substantiates this supposition. Using $J_{\text{at}} = +0.15 \text{ eV}$ leads to a contact contribution of only 2.5 G to the hyperfine interaction. The experimental value

of ΔA in Au:Yb is approximately 26 G, so that we attribute the remainder of 24 G to covalent mixing polarization and core polarization. Using (3) and our estimate of J_{cm} , we find a value for $A_{ce}^{cp}/g_I\mu_I = -4 \times 10^6$ G. Interestingly, this result, appropriate to admixtures of predominantly f -like conduction-electron states, is close to that value obtained by Gossard *et al.*¹ for the core-polarization contribution arising from d conduction-electron states for Yb metal (they quote a value of -2.4×10^6 G). However, it exceeds, by a factor of 4, estimates made by Crecelius and Hufner²³ on Eu dissolved in Ca and Yb metal. The great uncertainties which enter into all three estimations make comparisons rather meaningless beyond noting that the same or-

der of magnitude does obtain for $A_{ce}^{cp}/g_I\mu_I$ in three quite different materials.

IV. CONCLUSION

We have observed the hyperfine splitting of Er and Yb in Au. We have demonstrated the existence of an additional positive contribution to the hyperfine constant originating with the conduction electrons. A correlation between theory and experiment enables us, for the first time, to separate the covalent mixing and atomic contributions to the total exchange interaction, and to determine the combined p and f core-polarization contribution to the hyperfine constant.

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¹⁸This conclusion is altered if the mixing potential is nonspherical. In addition, band hybridization can "blur" an atomiclike characterization of conduction-electron states viewed from the impurity site. Either complication can alter a quantitative description of the process, but will have little impact on our conclusions so long as the mixing terms do not contribute to the contact hyperfine interaction.

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²⁰The terms in Eq. (3) express the majority of the contributions to ΔA in a dilute alloy. In a more concentrated material or magnetic host, additional contributions to the hyperfine field obtain. For example, in GdFe₂, R. E. Gegenwarth, J. I. Budnick, S. Skalski, and J. H. Wernick [*Phys. Rev. Letters* **18**, 9 (1967)] find a positive increase of hyperfine field of over 800 kOe as compared to the insulator value. One can analyze this result by noting that they found a positive increase in the hyperfine field from 160 kOe for GdPt₂ to 237 kOe for GdAl₂, which could be caused by contributions akin to Eq. (3) of this paper; and arguing for a "direct" contribution to the hyperfine field from the aligned Fe site d electrons, as did Gegenwarth *et al.* in their letter. Yet additional mechanisms could be cited for this positive contribution to the hyperfine field in the concentrated ferromagnet (e.g., spin-polarized s band), so that any direct comparison with the results we quote in this paper may be very misleading.

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