

Crystal-field and spin-exchange parameters in Ag-Dy and Ag-Er†

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We report our measurements of the temperature dependence of the static susceptibility and electron spin resonances for the systems Ag-¹⁶⁴Dy and Ag-Er. For both systems we determine the ground state is Γ_7 separated from a Γ_8 excited state by $\Delta = 11.5 \pm 1$ K, and 30 ± 5 K, respectively. The overall multiplet splitting is 160 ± 25 K for both. In addition to unambiguously identifying the crystal-field parameters, the reflection and transmission electron-spin-resonance spectra are analyzed in terms of a $J(k, k')\vec{S} \cdot \vec{s}$ model. We find that the ratio of the appropriate averages of $J(k, k')$ as determined from the temperature dependence of the g -shift and linewidth data, $J_g/J_{\Delta H}$, to be $\simeq 2$ (for a free-electron density of states).

We report our measurements of the temperature dependence of the static susceptibility and electron spin resonances for the systems Ag-¹⁶⁴Dy and Ag-Er. Our motivation for determining the properties had several components. Schultz *et al.* in an investigation of the equations appropriate for a description of the spin dynamics of the coupled local-moment-conduction-electron system, demonstrated that the transmission-spin-resonance technique could be used to simultaneously observe two well-resolved modes in Ag-Er.¹ In their analysis they suggested that there might be difficulties with characterizing the spin-dynamical behavior by the $J\vec{S} \cdot \vec{s}$ model² and we sought additional analogous systems. The major feature of the Ag-Er system, which distinguishes it from many other dilute local moment systems that have been investigated, is the presence of two well-resolved, albeit coupled, resonances. One of these resonances is due to the conduction electrons (for pure silver, $g_s = 1.983$),³ and the other due to the Γ_7 ground state of the Er local moment in a cubic crystal field, with $g_f \simeq 6.8$.⁴ The ratio of the respective g values is very large compared to the more usual case with S-state ions such as Mn, Cr, or Gd, where the analogous g -value ratio is essentially unity. As the dynamical equations governing the behavior of the local moment in the metal host have terms which are strongly dependent upon the ratio of the g values for the two spin species, it was felt desirable to be able to experimentally check the dependence on this parameter. The observation of electron spin resonance for the Ag-Dy system with a g value of 7.6 had been reported in the literature⁵ and thus seemed

particularly attractive for further study as it had the same host as Ag-Er.

Siebert *et al.*,⁶ reporting on their resonant measurements in Al-Er alloys, demonstrated the utility of ESR data for determining crystal-field parameters, and in particular pointed out the importance of including the effects of higher multiplet levels in the dc susceptibility.² Since the modification to the dc susceptibility is appreciable at the low temperatures utilized it was deemed most desirable to measure this property directly.

Finally, the choice of the Ag-Dy system seemed optimum for determining the crystal-field parameters with a significantly increased accuracy. Williams and Hirst suggested that there was a Γ_7 ground state but with a separation to the Γ_8 level of $\Delta \simeq 1$ K.⁷ Davidov *et al.*,⁵ in an analysis of their reflection electron-spin-resonance data for the $g \simeq 7.6$ line, concluded that $5 \leq \Delta \leq 10$ K. (After a reexamination of their analysis we suggested limits of $2 \leq \Delta \leq 20$ K.) Were the separation of the ground-state levels only ≈ 2 K, the Zeeman splittings of the multiplet could have been tuned over a range that would become experimentally accessible for possible direct observation of the transitions between the Γ_7 ground state and excited levels of the multiplet, and as we were interested in making such measurements, we needed to determine Δ more accurately.

We find that the combination of dc susceptibility, transmission spin resonance, and reflection spin resonance data (particularly at several frequencies) is a very powerful approach which allows for an unambiguous, accurate determination of crystal-field parameters. As shall be discussed,

all the data are consistent with theory with only one observation not yet explained (a large angular dependence of the reflection linewidth at 35 GHz) for Ag-Dy.

I. EXPERIMENTAL TECHNIQUES

The single crystals used were grown in vacuo by the Bridgeman technique. The silver was 69 grade Cominco. The ^{164}Dy was obtained via thoria reduction of the oxide. The Er used had the natural isotopic composition.⁸ A master alloy was first prepared and then, via dilution, crystals grown at stoichiometric concentrations between 20–200 ppm (by weight). The concentrations were checked by atomic absorption spectroscopy and (for the samples indicated) by dc susceptibility. The homogeneity of successive slices of oriented boules was checked by determining the resistivity at 4.2 K. The concentration accuracy is $\approx \pm 10\%$.

The susceptibility data were obtained utilizing the Faraday method.⁹ The reflection ESR data were obtained utilizing superheterodyne spectrometers at both 9 and 35 GHz. The transmission ESR data were obtained utilizing a transmission spectrometer at 9.2 GHz, whose performance was considerably improved by the addition of a maser preamplifier.¹⁰ The transmission samples were prepared by polishing oriented slices of the single crystals down to a thickness of ≈ 0.02 mm. All three spectrometers are provided with suitable magnets for producing and sweeping a dc magnetic field which could be rotated in a plane normal to the surface of the sample. The temperature of the sample was either determined from the vapor pressure of the surrounding liquid, or from suitable calibrated resistors.

II. DATA

A. dc susceptibility

The magnetization of a given sample was measured at selected fixed temperatures from 0.4 to 35 K as a function of dc fields up to 10 kG. These data were then corrected for the known diamagnetic susceptibility of pure Ag.¹¹ The magnetization data points, so corrected, for a Ag- ^{164}Dy sample are shown as the square points in Fig. 1. As can be seen, the magnetization does not saturate, but approaches a linear field dependence above ≈ 6 kG. The construction indicated was used to separate the total magnetization (M_T) into that part attributable to an isolated doublet ground state with a g value of 7.55 and that part due to the mixing of the ground state with the excited states of the multiplet (M_{VV}), i.e., the well-known Van Vleck contribution. The calculated curves shown in Fig. 1 will be discussed later in the analysis section.

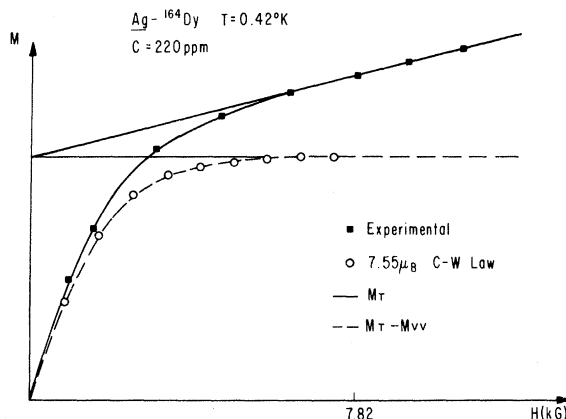


FIG. 1. Magnetization of a 220-ppm Ag- ^{164}Dy alloy as a function of magnetic field at $T=0.42$ K. The solid curve which is a best fit to the data (M_T) represents a Curie-Weiss law magnetization for a doublet with a magnetic moment of $7.55 \times \frac{1}{2} \mu_B$ (the dashed curve) plus a Van Vleck contribution (M_{VV}) as discussed in Sec. III. The Curie-Weiss temperature is $\theta = -0.1$ K. The error of the data points is $\pm 3\%$.

In Fig. 2 we have plotted the reciprocal of the initial susceptibility as a function of temperature for the same sample as in Fig. 1. The deviation from a $1/T$ law is clearly seen. In the computer analysis we have shown that the deviations are entirely due to the M_{VV} contribution, and not ordering, as the Curie-Weiss temperature is negligible ($\Theta = -0.1$ K).

Figures 3 and 4 are for data and analysis of a Ag-Er sample analogous to Figs. 1 and 2, respectively.

B. Transmission spin resonance

In Fig. 5 we present a typical spectrum of the transmission electron spin resonance (TESR)

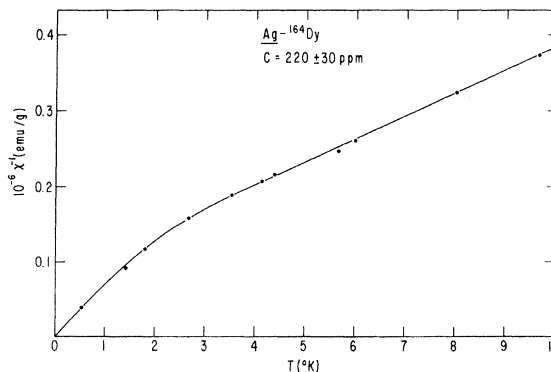


FIG. 2. The reciprocal of the initial susceptibility as a function of temperature for a 220-ppm Ag- ^{164}Dy alloy. The deviation from a $1/T$ law is clearly seen.

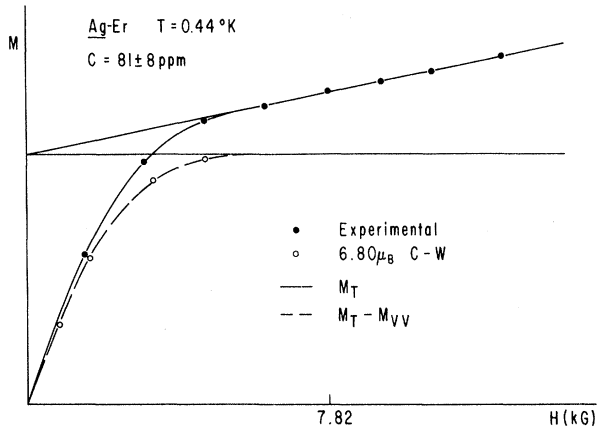


FIG. 3. The magnetization of an 81-ppm $Ag-Er$ alloy as a function of magnetic field at $T=0.44$ K. The solid curve which is a best fit to the data (M_T) represents a Curie-Weiss law magnetization for a doublet with a magnetic moment of $6.80 \times \frac{1}{2} \mu_B$ (the dashed curve) plus a Van Vleck contribution (M_{VV}) as discussed in the analysis. The Curie-Weiss temperature $\theta = -0.07$ K. The error of the data points is $\pm 3\%$.

signal showing the two resonances. The ordinate represents the component of the transmitted rf field that is in phase with a reference field adjusted (in this case) so as to yield an antisymmetrical low field resonance. The primary data for the low field resonance is the g value, which we find to be 7.66 ± 0.03 , and independent of the orientation of the dc field within this accuracy. This line is very quickly attenuated with increasing temperature, and can only be followed up to ≈ 2.5 K, in contrast to the reflection measurements discussed in Sec. IIC.

The high field line shown in Fig. 5 is strongly

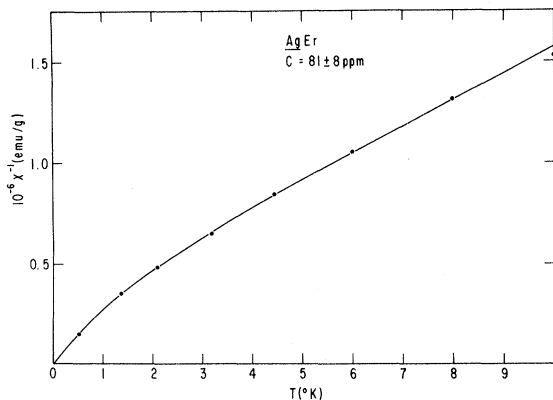


FIG. 4. The reciprocal of the initial susceptibility as a function of temperature for an 81-ppm $Ag-Er$ alloy. The deviation from a $1/T$ law are clearly seen.

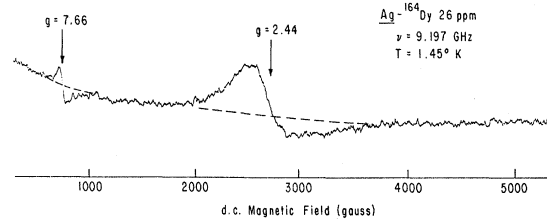


FIG. 5. Typical recorder trace of transmission signal as a function of the applied dc magnetic field at 1.45 K. The signal is defined as the component of the transmitted microwave field projected on a reference field. For the trace shown the phase of the reference has been chosen so as to display an antisymmetric low field signal. The low field resonance at $g=7.66$ is predominantly that of the Dy local moment corresponding to a Γ_7 doublet ground state. The high field resonance is due to the conduction electrons as g shifted from the pure metal value by the effective field of the local moments. For the temperature shown the g value is 2.44 corresponding to a g shift from the pure metal of $\Delta g = 0.46$.

shifted from the g value of pure Ag (1.983), and we designate the experimental g shift, $\Delta g_2 = g(\text{alloy}) - g(\text{pure Ag})$. In Fig. 6 we present Δg_2 as a function of temperature for a $Ag-^{164}Dy$ sample. We have also plotted another experimental quantity, Δg_1 , taken to be proportional to the dc susceptibility. The interpretation and analysis of the proportionality constant will be discussed in the next section.

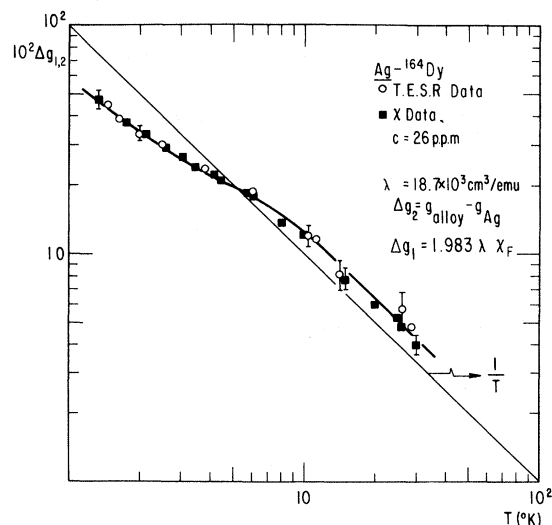


FIG. 6. The TESR data g shift, Δg_2 for the high field resonance in a 26-ppm $Ag-^{164}Dy$ alloy vs temperature. The g shift, Δg_1 , is taken as proportional to the dc susceptibility, with the constant given by 1.983λ as discussed in the analysis. The heavy line represents Δg_1 using the calculated susceptibility for $\Delta = 11.5$ K and $\alpha = 0.53$.

In Fig. 7 we have presented the (full) linewidth of the high field resonance as a function of temperature. In principle these data, in conjunction with the dc susceptibility, can be analyzed for the electronic spin-relaxation rate but, in practice, we find they are not sufficiently precise to yield useful results.

In Fig. 8 we present Δg_2 data for two Ag-Er alloys. The Δg_2 data for the 57-ppm sample are scaled to match the 81-ppm concentration. These Δg_2 data are the same as that presented in Fig. 2 of Ref. 1, but the sample concentration have been slightly corrected following our measurement of the susceptibility. The Δg_1 data clearly demonstrate that the g shift is proportional to the dc susceptibility as mentioned.^{2,6}

C. Reflection spin-resonance data

In the reflection mode only the low field line is observable due to signal-to-noise considerations, and it is the usual field derivative of the absorption that is actually measured. The line shapes display an admixture of the pure real and imaginary response due to the skin effect, and the data are analyzed assuming an intrinsic Lorentzian line.¹² The range of the data taken and their characterization is as follows.

In Fig. 9 we present the relaxation rate as a function of temperature 1.4–4 K, and at a frequency of 9.06 GHz. Neither the linewidth nor g value of this resonance exhibited any appreciable anisotropy as a function of orientation of the dc field to the sample at this frequency. Although we believe the relaxation rate is not a linear function of temperature, for reasons discussed below, these data are not sufficiently accurate to readily detect the anticipated deviations. Since it has

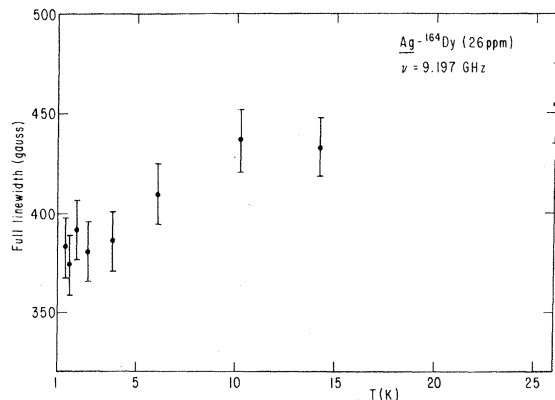


FIG. 7. The full linewidth, in gauss, of the high field TESR resonance for a 26-ppm Ag-¹⁶⁴Dy sample vs temperature at a frequency of 9.197 GHz.

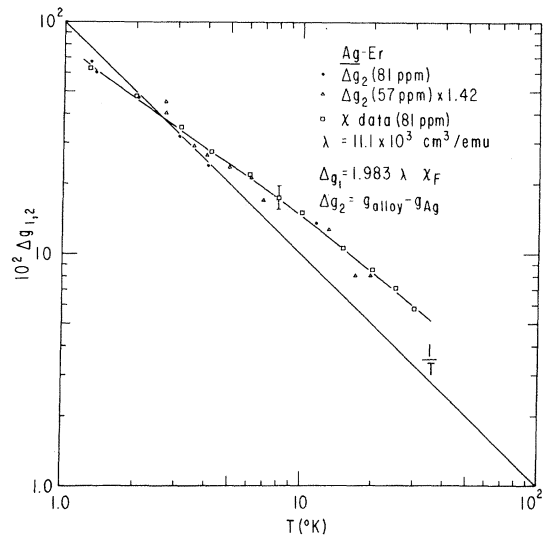


FIG. 8. The TESR g shift, Δg_2 , for the high field resonance in an 81-ppm Ag-Er alloy vs temperature. The analogous shift for the 57-ppm alloy is scaled by the concentration ratio. The g shift Δg_1 is taken as proportional to the dc susceptibility, with the constant given by 1.983λ as discussed in the analysis. The heavy line represents Δg_1 using the calculated susceptibility for $\Delta = 30$ K, and $\alpha = -0.33$.

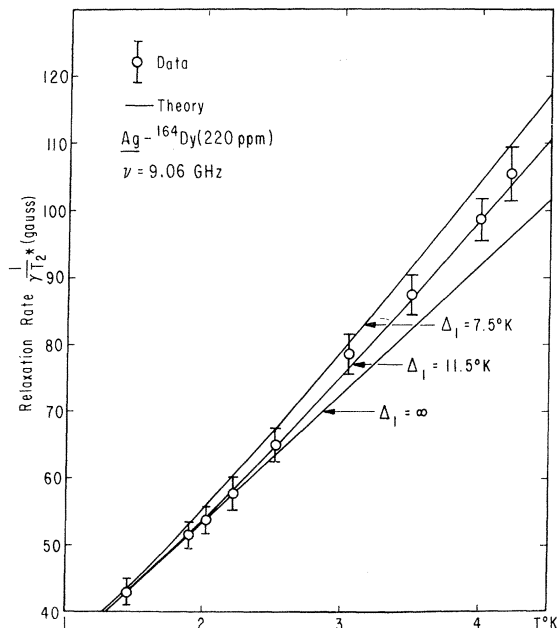


FIG. 9. The relaxation rate $1/\gamma T_2^*$ (in gauss) as determined from the low field reflection resonance linewidth vs temperature for a 220-ppm Ag-¹⁶⁴Dy alloy. The frequency was 9.06 GHz. The three theoretical curves correspond to $\alpha = 0.53$ and different choices of the ground state to first excited state splitting, Δ , and are calculated as discussed in the analysis.

been customary in the literature for such data to be analyzed in terms of a Korringa-like expression of the form $a + bT$, we note that the initial slope (corresponding to $\Delta = \infty$) is 18.5 ± 2 G/K.

Although we have remarked that there was negligible variation of the linewidth and g value of the low field resonance with magnetic field orientation at 9 GHz, this was decidedly not the case at 35 GHz. In Fig. 10 we present data of the g value of the low field line as a function of orientation of the applied dc magnetic field at a temperature of 1.4 K. The angular variation is consistent with that expected for a cubic environment as later discussed. We also observed appreciable anisotropy in the linewidth with variations at $\approx 15\%$ for which we have no explanation.

III. ANALYSIS

A. Susceptibility

We have analyzed the data of Figs. 1–4 in terms of a crystal-field split multiplet. The theoretical expressions used to compute the susceptibility (and other properties) are based upon a spin Hamiltonian for cubic symmetry which includes the crystal field to sixth order. We diagonalize the full Hamiltonian for a $J = \frac{15}{2}$ multiplet, including the Zeeman contributions, to get the appropriate wave functions and matrix elements. The dc susceptibility is calculated using the standard expression.¹³ Although the numerical results differ appreciably for the Dy and Er alloys, the methods of analysis are the same, and in what follows we shall discuss the Dy analysis in detail,

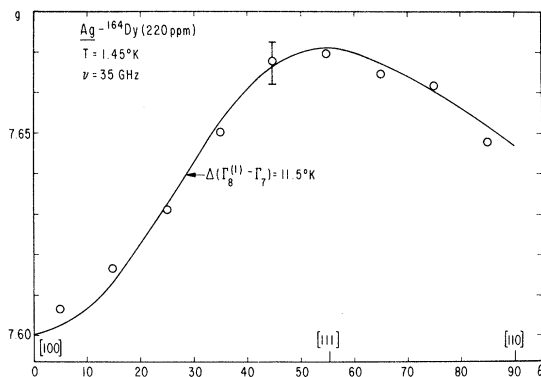


FIG. 10. The variation of the g value for the low field reflection resonance vs angular orientation of the applied dc field to the sample crystallographic axis at a frequency of 35 GHz. The normal to the crystal is $[110]$. The theoretical curve is calculated for a splitting $\Delta = 11.5$ K and $x = 0.53$. A constant g shift, corresponding to the exchange field, is added to the theoretical results as discussed in the text.

commenting upon the Er alloys only when necessary.

The Dy^{3+} ion is characterized by ${}^6H_{15/2}$. The observed low field ESR g value of 7.66 is very close to that for a pure Γ_7 doublet appropriate to the J , L , and S , (7.55). Our program calculates the susceptibility for a given concentration and trial values of Δ and x , reiterating to a least-squares fit to the data (x is in the notation of Lea, Leask, and Wolf).¹⁴ We actually find two regions which potentially fit the data, $\Delta \approx 10$ K, $x \approx 0.5$, and $\Delta \approx 30$ K, $x \approx -0.3$. Additional information is needed to make a unique assignment, and as we shall discuss in our analysis of the ESR data, the g anisotropy at 35 GHz can discriminate between the two. We find x positive for Ag-Dy, and our best fit to the data as shown in Figs. 1 and 2 correspond to $\Delta = 11.5 \pm 1$ K, $x = 0.53 \pm 0.01$, and an overall multiplet splitting of 160 ± 25 K.

If one compares the data presented in Figs. 2 and 4 one finds almost the identical temperature dependence. Thus the same two regions of Δ and x could fit the Ag-Er as well. Although we have not performed the ESR anisotropy experiments that could unambiguously confirm the appropriate designation, we believe that x for Er is negative (i.e., opposite to Dy), as is the case for these ions in insulators.¹⁵ The best fit to the data as presented in Figs. 3 and 4 correspond to $\Delta = 30 \pm 5$ K, $x = -0.33 \pm 0.02$, and an overall splitting of 160 ± 25 K.

We note that these results are in close agreement with those of Ref. 7 for Ag-Er, but our value of Δ is appreciably larger for Ag-Dy.

The computer program for susceptibility does not include the exchange field of the conduction electrons and hence the g value used to specify the size of the moment is the theoretical value for the Γ_7 doublet (as shown in Figs. 1 and 3) instead of the observed ESR g values. The deviations from these values are discussed below. The data are analyzed using the Curie-Weiss form for the basic susceptibility and we find very small Curie-Weiss temperatures Θ , indicating that for our concentrations and temperature range there is no problem with ordering. We find $\Theta = -0.1$ K, and -0.07 K for Ag-Dy and Ag-Er, respectively.

B. ESR and TESR

As was mentioned earlier, one of the primary motivations for making these detailed measurements on the Ag-Dy system was the results and analysis reported by Schultz *et al.* in Ag-Er.¹ After incorporating the effects of the crystal field one may ask, what are the dynamical equations appropriate for describing such local moments

in the metallic host? We analyze the ESR data within the framework of a $J(k, k')\vec{S} \cdot \vec{s}$ model as discussed in Ref. 1, and also recently for Ag-Gd.¹⁶ For the case considered here, the resonances are completely “unbottle-necked” by virtue of the large disparity in g values between the ions and the conduction electrons, and therefore the simple formula for the exchange fields may be used to calculate g shifts in the analysis that follows. (The results of the more elaborate computer calculations verify this situation.)

1. High field resonance g -shift data at 9 GHz

The host conduction-electron-spin resonance occurs at $g_s = 1.983$, but the observed high field resonance is shifted from this value by the exchange field of the local moments resulting in the shift $\Delta g_1 = 1.983\lambda\chi_F$, where χ_F is the dc susceptibility of the local moments, and λ is given by $J_g\Omega/g_s g_f \mu_B^2$. Here Ω is the atomic volume, g_s and g_f refer to the conduction electrons and local moments, respectively, and μ_B is the Bohr magneton. J_g represents the average of the exchange constant $J(k, k')$ appropriate to the g shift.¹⁷ The only major way crystal-field effects enter in this relation is that χ_F is not represented by a simple Curie law as discussed. In Figs. 6 and 8 we compare the experimental values of the g shift of the high field resonance, Δg_2 , with the susceptibility data, thereby obtaining a value for λ which sets the normalization. The values for λ are given in Figs. 6 and 8 while the corresponding J_g are presented in Table I.

2. Low field resonance g -shift data at 9 GHz

While the high field resonance is g shifted by the exchange field of the moments, the low field resonance is g shifted in an analogous manner with $\Delta g_f/g_f = \lambda\chi_s$, i.e., the Knight shift. Since χ_s , the conduction-electron susceptibility is virtually temperature independent we can get another value for λ by evaluating the g shift at the lowest temperature corresponding to the most accurate reflection g value measurements. We find $g_f = 7.66$

± 0.01 . The resulting accuracy for λ is comparable to our previous method, but it must also be borne in mind that there might be other mechanisms for slight shifts from the theoretical values which are specific to moments in metals, and which have not yet been discovered. Since this g shift is very small, such corrections would have a large effect on λ . Also χ_s should be the many-body enhanced susceptibility. Notwithstanding these reservations, we have calculated the corresponding values of J_g using χ_s calculated for a free electron gas, and taking $m^*/m = 1$. They are presented in Table I. The agreement of these values with those for J_g deduced from the $\Delta g_{1,2}$ data is very satisfactory.

3. Low field resonance linewidth data at 9 GHz

In the absence of crystal-field effects, the low field resonance relaxation rate (or linewidth) should be expressible in the form $a + bT$. From a study of the temperature dependence one may calculate the quantity $J_{\Delta H}$, which is the average of the exchange constant $J(k, k')$ appropriate to the linewidth.¹⁷ We have calculated the expected temperature dependence of the relaxation rate for Ag-Dy including crystal-field effects for various values of Δ , using values of wave functions and matrix elements from our diagonalization of the full Hamiltonian described in the susceptibility analysis, and incorporating the expression given by Hirst.¹⁸ These results are shown as the solid curves in Fig. 9. As can be seen, the data are consistent with a value of $\Delta = 11.5$ K, but it would be difficult to reverse the process to obtain an accurate value solely from the linewidth. The values of $J_{\Delta H}$ as deduced from the data are also presented in Table I.

4. Low field resonance g -anisotropy data at 35 GHz

The crystal field can result in appreciable g anisotropy with respect to the orientation of the dc field. Although the effect was not observable at 9 GHz, it was clearly present for the Ag-Dy alloy

TABLE I. Crystal-field and electron spin resonance parameters for dilute rare-earth: Ag alloys.

	Δ (K)	x	Overall splitting (K)	J_g^{TESR} (eV)	J_g^{RESR} (eV)	$J_{\Delta H}^{\text{RESR}}$ (eV)	$J_{\Delta g}/J_{\Delta H}$
				High-field line	Low-field line	Low-field line	
Dy	11.5 ± 1	0.53 ± 0.01	160 ± 25	0.89 ± 0.09	0.89 ± 0.1	0.42 ± 0.04	2.1 ± 0.3
Er	30 ± 5	-0.33 ± 0.02	160 ± 25	0.46 ± 0.06	0.44 ± 0.15	0.24 ± 0.03	1.9 ± 0.4
Gd ^a					0.76 ± 0.10	0.25 ± 0.025	3.0 ± 0.6

^a Reference 16.

at 35 GHz as presented in Fig. 10. We have calculated the g anisotropy to be expected at $x = 0.53$ and also -0.33 following the expression as given in Eq. (2) of Ref. 5. We find that while there is negligible variation, ($<0.1\%$), at $x = -0.33$, there is the large variation, ($\sim 1.3\%$) at $x = 0.53$, as presented in Fig. 10. (A constant g shift of 7.66–7.55 has been added to the theoretical results to incorporate the effects of the exchange field as discussed in Sec. III B 2.) Thus the g -anisotropy feature of the ESR data allows an unambiguous choice of the crystal-field parameters without the need to resort to theoretical model calculations for the sign of x . As mentioned, we have not made the corresponding determinations for Ag-Er, but would predict essentially no anisotropy corresponding to the $x = -0.33$ results.

5. Low field resonance linewidth anisotropy data at 35 GHz

Along with the g anisotropy just discussed, we observed substantial variations, $\approx 15\%$, in the linewidth of the Ag-Dy alloy at 35 GHz. While our computer results predict the correct angular dependence of this variation, the magnitude is much too small, only $\approx 0.5\%$. We currently have no explanation for the source of this variation, and can only suggest strain or related effects.

IV. DISCUSSION

The principal results relating to crystal field and ESR parameters have been presented in Table I, where, for purposes of comparison, we have also included the exchange parameters for Ag-Gd.

As mentioned at the outset, part of our initial motivation for working with the Ag-Dy system was the results and analysis that had been presented for Ag-Er.¹ In that work the authors attempted to interpret their g -shift data within the framework of a $J(k, k')\vec{S} \cdot \vec{S}$ model, amongst others, but as noted in Ref. 2, erroneously calculated the temperature dependence of the susceptibility as a simple Curie law and neglected the Van Vleck contribution. In fitting the lowest-temperature data, they obtained a ratio $J_{\Delta g}/J_{\Delta H} \approx 3$. As we have shown in Fig. 8, the experimental g -shift data is quite accurately proportional to the (measured) dc susceptibility, and as is presented in Table I, the revised analysis results in $J_{\Delta g}/J_{\Delta H} \approx 1.9$, very close to the value for Ag-Dy. We note that the ratio for Ag-Gd is ≈ 3 , and, whether one regards these values as a serious challenge to the meaningfulness of the $J(k, k')$ expansion in terms of Legendre coefficients, is currently a matter of perspective.¹⁹ We believe that these data form a useful set, and suggest that what is now needed are theoretical calculations of these parameters to test models of the local moment environment, and to provide direction for further experimental effort.

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²The analysis used in Ref. 1 suggested that $J_g/J_{\Delta H} \approx 3$. As pointed out in Ref. 6 below, that analysis neglected the Van Vleck contribution to the susceptibility, and consequently $J_g/J_{\Delta H}$ is really ≈ 2 . These data are further reviewed in the Sec. IV of this paper.

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plications due to the hyperfine interaction. In Ag-Er one observes a very strong dominant line even with the naturally occurring isotopic composition.

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¹⁹Perspective also includes an element of orientation,

or set. The suggestion made in Ref. 1, that the g -shift data might be best interpreted as a constant J with a temperature-dependent g value (corresponding in turn to temperature-independent g value in the representation of their case B), was appealing at that time for two reasons: (i) it gave a reasonable fit to the data, and (ii) the authors were seeking some physical criteria that might naturally suggest canonical M_s - M_d representation for which to analyze the local moment problem, as opposed to the infinity of representations which they had just shown were mathematically allowed. Whether there is a particular natural representation still remains an interesting problem.