solved in the limit of M, n large by methods similar to those used for the normal Bethe *Ansatz*.[13,14] Using (22), (18), and (9) we then obtain the result (2).

We intend to give details elsewhere. We remark that $k_1, \ldots, k_n$ are real and distributed symmetrically over the interval $(-\frac{1}{2}\pi, \frac{1}{2}\pi)$, that $n = M$, and that $E_1, \ldots, E_n$ are real and non-negative.

One of us (F.Y.W.) wishes to thank Professor K. J. Le Couteur for his kind hospitality at the Australian National University.

Added note.—Numerical estimates of the critical indices α', β, and γ' of the present model have recently been given by H. P. Griffiths and D. W. Wood, J. Phys. C: Proc. Phys. Soc., London 6, 2533 (1973).

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Clarification of Local-Moment–Conduction-Electron Resonance*

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Our data on spin resonances in *Ag*-Er are inconsistent with the $J\vec{S}\cdot\vec{s}$ model, and barely consistent with $J(\hat{k},\hat{k}')$. Alternatively, a suggestive temperature dependence of the $J\vec{S}\cdot\vec{s}$ conduction-electron g value is compatible with the data. We present general kinetic equations for a local-moment–conduction-electron system. We clarify questions of detailed balance, relaxation to exchange fields, positive energy absorption, and reduction of the kinetic equations for any possible model to either of two standard forms.

We present results for the electron spin-resonance parameters for the local-moment–conduction-electron spin system *Ag*-Er. This system is characterized by two widely separated resonances (the g value for the conduction electrons is $\simeq 2.0$, while for Er, $g \simeq 6.8$), which are coupled via an exchange interaction. In the course of our efforts to analyze this system we have completed a theoretical and computational study which we believe resolves important questions, and raises new ones, concerning the dynamical description of a dilute local moment in a metal.

The experiments were performed utilizing the transmission electron spin-resonance (TESR) technique.[1] The samples were typically single-crystal foils 0.002 cm thick containing between 20 and 70 ppm of Er in Ag.[2] The sample temperature was varied between 1.25–30°K. The signal in the TESR technique is defined as the projection of the transmitted microwave field on a reference field of the same frequency. The data are taken at fixed temperature, frequency, and reference phase angle, while the applied dc field is slowly swept through the resonance conditions.

In Fig. 1 we present such a transmitted signal versus dc field where the phase of the reference has been chosen so as to yield a symmetric high-field resonance (HFR). The low-field resonance (LFR) is clearly seen and for the particular set of conditions also appears symmetric.[3] The general properties of the resonances are the following: The LFR amplitude ($g \cong 6.84$) is slightly larger than the HFR at 1.3°K. The LFR broadens linearly with temperature, and the amplitude decreases somewhat faster than $1/T^2$. Consequently, it was only observable up to temperature $\simeq 3°\text{K}$. The HFR ($g \cong 2-2.6$) broadens slightly with temperature, and the amplitude decreases fairly slowly and was observable up to temperature $\simeq 30°\text{K}$. The g value of the LFR varied very slightly with temperature while that of the HFR varied substantially as discussed below. Accurate simultaneous measurements on both resonances in a single sample make the following analysis possible.

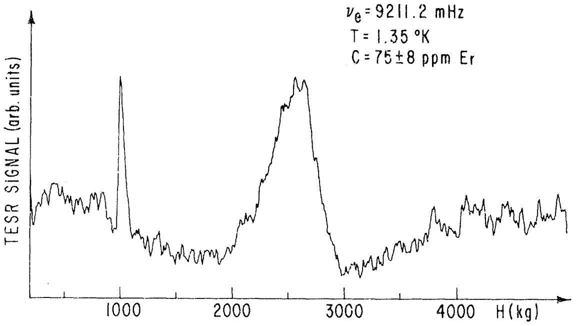

FIG. 1. The signal (that component of the transmitted microwave field which yields a symmetric line shape) versus the applied dc magnetic field. The low-field resonance g value is 6.84 ± 0.02; the high-field resonance g valus is 2.59 ± 0.05.

We write the equations for the conduction-electron (s) magnetization and the local-moment (f) magnetization, where we specifically allow for different g values, as

$$\frac{\partial \vec{\mathrm{M}}_s}{\partial t} = \gamma_s(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_f + \lambda_s \vec{\mathrm{M}}_f) \times \vec{\mathrm{M}}_s + \frac{\gamma_s}{\gamma_f T_{fs}}[\vec{\mathrm{M}}_f - \chi_{f0}(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_s)] - \left(\frac{1}{T_{sf}} + \frac{1}{T_{sL}}\right)[\vec{\mathrm{M}}_s - \chi_{s0}(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_f)]$$
$$+ D\nabla^2[\vec{\mathrm{M}}_s - \chi_{s0}(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_f)], \qquad (1)$$

$$\frac{\partial \vec{\mathrm{M}}_f}{\partial t} = \gamma_f(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_s + \lambda_f \vec{\mathrm{M}}_s) \times \vec{\mathrm{M}}_f + \frac{\gamma_f}{\gamma_s T_{sf}}[\vec{\mathrm{M}}_s - \chi_{s0}(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_f)] - \left(\frac{1}{T_{fs}} + \frac{1}{T_{fL}}\right)[\vec{\mathrm{M}}_f - \chi_{f0}(\vec{\mathrm{H}} + \lambda \vec{\mathrm{M}}_s)],$$

with the boundary condition at the sample surface $\partial \vec{\mathrm{M}}_s/\partial n = -\alpha[\vec{\mathrm{M}}_s - \chi_{f0}(\vec{\mathrm{H}} + \lambda M_s)]$, where it is intended that (1) shall be linearized about equilibrium in the dc field $\vec{\mathrm{H}}_0 = H_0\hat{z}$. In the resulting equations for $M_{s,f}^{x,y}$, $\chi_{s0} = M_s^z/(H_0 + \lambda M_f^z)$ and $\chi_{f0} = M_f^z/(H_0 + \lambda M_s^z)$. As a consequence of time-reversal invariance, $T_{fs}/T_{sf} = \gamma_s^2 \chi_{f0}/\gamma_f^2 \chi_{s0}$, and $\gamma_s \lambda_s = \gamma_f \lambda_f$. The condition that the sample absorb energy implies $1/T_{sL}T_{fL} + 1/T_{sL}T_{fs} + 1/T_{fL}T_{sf} \geq 0$, $D \geq 0$, $\alpha \geq 0$. Subject to these restrictions, Eqs. (1) are the *most general possible* if all coefficients are permitted to be functions of H_0, the temperature, and the frequency ω, and are invariant under $\omega \to -\omega$, $H_0 \to -H_0$.[4] Any dependence on ω is irrelevant to the analysis of the present experiments, in which ω was fixed.

We now focus on two apparently contradictory cases which have been most discussed in the development of the resonance problem:

Form (A): $\lambda_s = \lambda_f = 0$.—This form occurs naturally for the $J\vec{\mathrm{S}} \cdot \vec{\mathrm{s}}$ model.[5] The microscopic model contains parameters γ_s^A, γ_f^A, $J(=\gamma_s\gamma_f\lambda/\Omega_0)$, χ_{s0} (Pauli susceptibility), T_{sL}^A, and D. γ_s^A, γ_f^A, and J are regarded as temperature independent. χ_{f0}, T_{sf}^A, and T_{fs}^A are calculated explicitly (e.g., T_{fs}^A is given by the usual Korringa formula). There is no theory at present for T_{fL}^A or α. We set $\alpha = 0$ assuming no surface spin relaxation.

Form (B): $\lambda = 0$.—This form occurs naturally for the Anderson model if the conduction-electron density of states ρ is regarded as constant. (The compensation theorem implies $\lambda^B = 0$.) A complicated analysis[6] yields the parameters of Eqs. (1) (except for T_{fL}^B and α), in terms of the parameters in the Anderson Hamiltonian; γ_s^B and γ_f^B are not equal to the microscopic values unless $\gamma_s^M = \gamma_f^M$ [$\gamma_{s,f}^M = (\epsilon_{s,f\uparrow} - \epsilon_{s,f\downarrow})/\hbar H_0$].

Even when $\gamma_s = \gamma_d$ (a circumstance which is usually discussed in connection with "bottlenecked" conditions), the kinetic equations for forms A and B appear to be strikingly different. Whereas both equations contain the exchange field in the torque terms, only form A includes the exchange field in the relaxation terms. Also note that unless $\gamma_s^B = \gamma_f^B$, there is an asymmetry in the exchange field.[7] A valuable advance was made towards the resolution of this paradox by Langreth and Wilkins[5] (see in particular their note in proof) who examined the nature of their equations for form A under the Schrieffer-Wolff transformation. They showed that the operators whose averages we denote by $\vec{\mathrm{M}}_s$ and $\vec{\mathrm{M}}_f$ are different in the two models, that the unitary transformation which reduces the Anderson model to the $J\vec{\mathrm{S}} \cdot \vec{\mathrm{s}}$ model does not leave $\vec{\mathrm{M}}_s$ and $\vec{\mathrm{M}}_f$ invariant, but rather induces a linear transformation in $\vec{\mathrm{M}}_s, \vec{\mathrm{M}}_f$ space, and that, for $\gamma_s = \gamma_f$, the equations of form B transform exactly to those of form A under this transformation. We have extended this linear transformation of $\vec{M}_s, \vec{M}_f$ to the case $\gamma_s \neq \gamma_f$, using a model-independent approach,[4] and we find that (1) can always be reduced to form A or B at will. We find generally that

$$\vec{M}_s{}^B = \vec{M}_s{}^A - \lambda\chi_{s0}\vec{M}_f{}^A,$$
$$\vec{M}_f{}^B = (1 + \lambda\chi_{s0})\vec{M}_f{}^A, \qquad (2)$$

and if Eqs. (1) are written in form A, and the transformation (2) is made, one obtains (1) in form B *with a new set of coefficients* which can be expressed in terms of the original (form A) set. [Note that $\vec{M} = \vec{M}_s + \vec{M}_f$ is invariant under (2), so that Maxwell's equations, which involve only the total magnetization density $\vec{M}$, are not modified.] As examples of the relationship between parameters we note $\gamma_s{}^B = \gamma_s{}^A + \lambda^A\chi_f{}^A(\gamma_s{}^A - \gamma_f{}^A)$, $\gamma_f{}^B = \gamma_f{}^A$, $\lambda_f{}^B = \lambda^A$, $\lambda_s{}^B = \lambda^A\gamma_f{}^B/\gamma_s{}^B$ $[\chi_f{}^A = (M_f{}^A)_z/H_0]$.

We have calculated the transmitted field, using Maxwell's equations and (1), for form A where we evaluate all coefficients using the $J\vec{S}\cdot\vec{s}$ model, assuming $J(\hat{k},\hat{k}')$ is constant. We assume a value for $g_f (= 6.82)$. g_s for pure silver is known to be 1.983.[8] χ_{f0} is calculated from a Brillouin function using g_f, $S = \frac{1}{2}$, and the known Er concentration, C. Free-electron values are used for χ_{s0} and D. The results are qualitatively insensitive to the values used for χ_{s0}, D, and T_{sL}. λ, T_{sf}, and T_{fs} are expressed in terms of J (appropriately modified when $\hbar\gamma_f H_0/kT \geq 1$).[9] We find that (a) the LFR linewidth broadens linearly with temperature (as expected from a simple Korringa consideration); (b) the HFR g value shifts with temperature, reflecting the exchange field $\lambda^A M_f$. $(g - g_s)/JC$ is a universal function of temperature χ_f/C in this model. We have plotted this function in Fig. 2. The data are analyzed in a similar manner. The slope of the LFR linewidth versus temperature yields a value for $|J|$. We find $J = 0.23 \pm 0.015$ eV.[9] Knowing J (the sign is determined from the sign of the g shift) and C, we calculate $(g_{\text{obs}} - g_s)/JC$ for the HFR. These data are presented in Fig. 2. The discrepancy is striking and well beyond the error limits.

We have attempted to account for this discrepancy by allowing J to be a function of $\hat{k}\cdot\hat{k}'$, $J = \sum_l (2l+1)P_l(\vec{k}\cdot\vec{k})J_l$.[10] We find that (1) the data cannot be fitted *at all* by approximating the series with either J_0 and J_1, or J_0 and J_2; (2) for J_0 and J_3 is a fit to the data (within the error limits) is mathematically possible, but would require a questionably large *positive* value for $J_3 (= +0.4J_0)$. As an alternative approach, we have calculated $(g - g_s)/JC$ for the HFR by choosing $\gamma_s{}^A$ to be *dependent* on temperature in just such a way that $\gamma_s{}^B$ is *independent* of temperature (keeping J constant so that λ^A varies with temperature). The results in the dashed curve shown in Fig. 2.

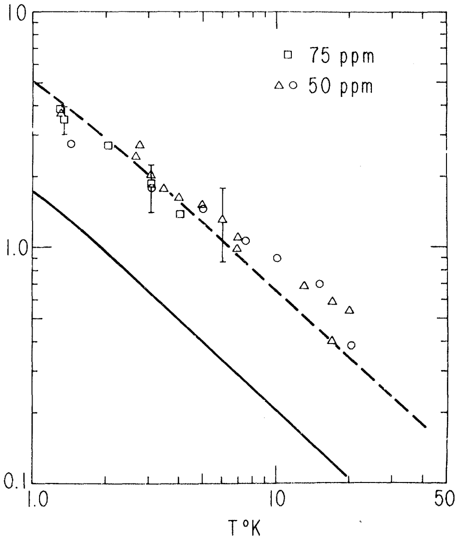

FIG. 2. We define the g shift, Δg, as g(observed or calculated for HFR) $-g$(pure silver). We have plotted $100\Delta g/JC$ versus temperature. J is obtained from an analysis of the slope of the low-field resonance linewidth versus temperature data. C is the Er concentration (atomic ppm). We find $J = 0.23 \pm 0.015$ eV. The heavy line corresponds to both the $J\vec{S}\cdot\vec{s}$ and Anderson model results as normally interpreted. The dashed line represents the results of calculations assuming a particular temperature dependence for the effective conduction-electron g value as discussed in the text. The increasing percentage error with temperature reflects a constant error in Δg of ± 0.05. The susceptibility used to calculate both the theoretical curves should be corrected for the admixture to the Γ_7 doublet above ≈ 5°K; this correction would bend the curves upward at higher temperatures.

To summarize our theoretical framework: (1) Equations (1) are completely general and model independent, and furnish a useful meeting ground for theory and experiment.[4] (2) Form A or B of (1) is equally valid, if the coefficients are given a suitable dependence on temperature; the choice is largely a question of convenience. The complete transformation of coefficients will be published elsewhere.[4] (3) The crucial question is not the choice of form A or B (i.e., whether or not the relaxation destination includes the exchange field), but rather the temperature dependence of the coefficients in (1).

To summarize our interpretation of the data for the *Ag*-Er system: (1) An analysis in terms of a wave-vector–dependent J for the $J\vec{S}\cdot\vec{s}$ model requires a disturbingly strong dependence on the scattering angle $\hat{k}\cdot\hat{k}'$. (2) Alternatively our data are consistent with a temperature dependence of γ_s^A such that γ_s^B is essentially constant.

Neither the $J\vec{S}\cdot\vec{s}$ nor the Anderson model predicts that γ_s^B is independent of temperature, but should further experimental evidence support this approach it would suggest that equations of form **B** may be conceptually closer to the correct microscopic theory of local moments.[11]

We acknowledge with pleasure conversations with Professor R. Orbach, Professor J. Wilkins, Professor A. Ron, Professor M. Fibish, Dr. Y. Yafet, Dr. D. Davidov, and Dr. J. Zitkova-Wilcox. We are indebted to Professor D. Wohlleben, Mr. B. Sales, and Dr. J. Huber for generously measuring the susceptibility. We thank Dr. S. Oseroff and Mr. C. Latham for their help, and Mr. J. Armstrong for his enthusiastic writing and execution of the computer programs. We also thank the San Diego Division of the Academic Senate of the University of California for a computer grant.

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[1] A discussion of our TESR apparatus may be found in *Metals*, edited by R. F. Bunshah (Wiley, New York, 1972), Vol. 16, Part 1, Chap. 5, and references cited therein.

[2] Determination of the Er concentration is a most difficult problem. We have utilized emission spectroscopy, resistivity ratio, dc susceptibility, neutron activation, and atomic absorption. The accuracy is estimated as ±10%.

[3] In general the LFR is not in phase with the HFR.

[4] D. R. Fredkin, to be published.

[5] Space prohibits listing all references. See J. Dupraz *et al.*, *Magnetic Resonance* (Plenum, New York, 1970), p. 192, for a useful review. More recently we note D. C. Langreth and J. W. Wilkins, Phys. Rev. 6, 3189 (1972); S. Barnes and J. Zitkova-Wilcox, Phys. Rev. B 7, 2163 (1973); W. Götze and P. Wölfle, J. Low Temp. Phys. 6, 455 (1972).

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[9] Neither J nor ρ appears separately in the formula for the signal, but rather the combination ρJ. All J values in the text are those for a free electron density of states.

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Quadrupole Exciton-Phonon Dynamics at the 151-K Phase Transition in $PrAlO_3$

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The dynamics of the 151-K Jahn-Teller phase transition in $PrAlO_3$ have been studied by means of inelastic neutron scattering; the transition is found to involve a soft $T[101]$ acoustic phonon which in turn is driven soft by a *quadrupole exciton mode*, an electronic mode involving a wavelike modulation of the Pr^{3+} electric quadrupole moment. We report measurements both of the soft phonon behavior and of the coupled excitations in the crossing region.

The perovskite $PrAlO_3$ is known to exhibit a cubic-rhombohedral (D_{3d}^6) soft Γ_{25} R-point phonon transition at 1320 K followed by a first-order transition to an orthorhombic phase (C_{2h}^3) at 205 K and a second-order transition at 151 K which at completion leaves the crystal essentially te-