Kondo effect is drastically reduced.

Very recently, Theumann[6] has published a study of the Anderson model which employed an essentially identical decoupling scheme to that used by AP for the Wolff model. Theumann treated the case where v was infinite and the Anderson mixing term V was small compared to the deep impurity level E_D. She first specialized to the case of a very deep impurity level. In this case, she found behavior quite similar to that found by Bloomfield and Hamann[7] for the s-d model, if the results for the resistivity were expressed in terms of T/T_K, where T_K, the Kondo temperature, was $T_K = De^{-E_D/\rho V^2}/\pi$, where D was the bandwidth, ρ was the density of conduction electrons, and V was the mixing term.

Our result essentially complements Theumann's by considering the strong-coupling Wolff-model case. It also contains the difficulty briefly discussed by Theumann, that not all the Kondo-like "leading logarithmic terms" are contained in the decoupling scheme.

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Magnon-Coupled Nuclear Spin-Phonon Interaction*

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The magnon-coupled nuclear spin-phonon interaction first proposed by Silverstein is discussed on the basis of a classical model. In addition to being conceptually quite simple, the model points out several interesting new features of this coupling mechanism.

The magnon-coupled nuclear spin-phonon interaction first proposed by Silverstein[1] has been observed in at least one antiferromagnet[2] and probably in several others.[3-5] The purpose of this paper is to discuss this coupling mechanism from a point of view different from that of Silverstein.

Following Silverstein[1] we consider a uniaxial two-sublattice antiferromagnet in which the nuclear spin system is coupled to the electronic spins via an isotropic hyperfine interaction. (A good example is the Mn^{55} nuclear spin system in MnF_2.) The Hamiltonian of such a system consists of the following terms:

$$\mathcal{H}=\mathcal{H}_{\mathrm{P}}+\mathcal{H}_{\mathrm{MP}}+\mathcal{H}_{\mathrm{ex}}+\mathcal{H}_{\mathrm{A}}+\mathcal{H}_{\mathrm{ZM}}+\mathcal{H}_{\mathrm{hyp}}+\mathcal{H}_{\mathrm{ZN}} \ . \qquad (1)$$

The individual terms correspond to the phonon, the magnon-phonon, the exchange, the magnetic anisotropy, the electronic Zeeman, the hyperfine, and the nuclear Zeeman contributions to the total Hamiltonian. Silverstein's solution to the problem involved expressing each term of Eq. (1) in terms of phonon and spin-wave operators, performing an average over the intermediate electronic spin system, and thus deriving an effective nuclear spin-phonon interaction, which is then used to calculate an ultrasonic absorption coefficient. This two-step process is described as follows: An acoustic phonon interacts with the electronic spin system (via $\mathcal{H}_{\mathrm{MP}}$) creating a virtual magnon; the magnon is then coupled to the nuclear spin system and produces a nuclear spin transition via the hyperfine interaction $\mathcal{H}_{\mathrm{hyp}}$. The calculation is clearly restricted to low temperatures ($T \ll T_N =$ Néel temperature) and, because of the average over the electronic spin states, any time correlation existing between the electronic and nuclear spins is lost.

An alternative approach to the problem is to calculate the elementary excitation spectrum of the complete system represented by the Hamiltonian of Eq. (1). Since the average over the electronic spin states is then unnecessary, time correlation effects between the two spin systems are automatically included. The rest of this paper is devoted to a completely classical calculation of this alternative approach.

The free-energy density of the system described by Eq. (1), given in terms of the macroscopic electronic sublattice magnetization vectors $\vec{M}_1$ and $\vec{M}_2$ ($|\vec{M}_1| = |\vec{M}_2| \equiv M_0$) and the nuclear sublattice magnetization vectors $\vec{I}_1$ and $\vec{I}_2$, may be written

$$E = E_P + M_0^{-1} b[(M_{1x} - M_{2x})e_{xz} + M_{1y} - M_{2y})e_{yz}] - \lambda \vec{M}_1 \cdot \vec{M}_2 - \tfrac{1}{2} M_0^{-2} K[M_{1z}^2 + M_{2z}^2] - \vec{H}_0 \cdot (\vec{M}_1 + \vec{M}_2) + A(\vec{M}_1 \cdot I_1 + \vec{M}_2 \cdot \vec{I}_2) - \vec{H}_0 \cdot (\vec{I}_1 + \vec{I}_2) \quad . \tag{2}$$

The phonon term E_P is a function of the adiabatic elastic constants c_{ij} and the elastic strains e_{ij}. For D_{4h}^{14} symmetry (e.g., for MnF_2) we have

$$E_P = \tfrac{1}{2} c_{11}(e_{xx}^2 + e_{yy}^2) + \tfrac{1}{2} c_{33} e_{zz}^2 + \tfrac{1}{2} c_{44}(e_{xz}^2 + e_{yz}^2) + \tfrac{1}{2} c_{66} e_{xy}^2 + c_{12} e_{xx} e_{yy} + c_{13}(e_{xx} e_{zz} + e_{yy} e_{zz}) . \tag{3}$$

b is the magnetoelastic coupling constant,[6] λ is the exchange constant, K is the magnetic anisotropy constant, A is the dimensionless hyperfine constant, and $\vec{H}_0$ is the applied dc magnetic field. The strains e_{ij} are defined in terms of the elastic displacement field $\vec{u}(\vec{r}, t)$ by

$$e_{ij} = \left(1 - \frac{\delta_{ij}}{2}\right)\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \quad . \tag{4}$$

The equations of motion for the elastic displacements[7] are

$$\rho \frac{\partial^2 u_x}{\partial t^2} = \frac{\partial^2 E}{\partial x \partial e_{xx}} + \frac{\partial^2 E}{\partial y \partial e_{xy}} + \frac{\partial^2 E}{\partial z \partial e_{xz}} \tag{5}$$

with cyclic permutations of x, y, z, for u_y and u_z. The equations for the magnetization components are

$$\frac{\partial M_{ij}}{\partial t} = \gamma_e \left[\vec{M}_i x \left(\frac{-\partial E}{\partial \vec{M}_i}\right)\right]_j \quad ,$$
$$\frac{\partial I_{ij}}{\partial t} = \gamma_N \left[\vec{I}_i x \left(\frac{-\partial E}{\partial \vec{I}_i}\right)\right]_j \quad , \tag{6}$$

with $i = 1, 2$ and $j = x, y$. Here γ_e and γ_N are the gyromagnetic ratios for the electronic and nuclear spin systems, respectively. After linearizing these equations and assuming harmonic variation in space and time, we obtain a secular equation, the solutions of which are the dispersion relations for the coupled phonon-magnon nuclear spin modes. Those solutions, which in the absence of magnetoelastic coupling correspond to purely elastic modes, can be written as effective or measured elastic constants. For the case of $\vec{H}_0 = 0$ we find[8]

$$c_{44}^* = c_{44} - 2\frac{b^2}{K} - 2\left(\frac{AbM_0}{K}\right)^2 \chi_N(T)\left(\frac{\omega^2}{\omega_H^2 - \omega^2}\right) . \tag{7}$$

The second term on the right-hand side of this equation (discussed more completely in Ref. 6) is the result of magnetoelastic coupling to the magnon modes in the limit that the lowest-frequency magnon mode is much higher than the acoustic frequency. The last term is the dispersion associated with the magnon-coupled nuclear spin-phonon interaction. $\chi_N(T)$ is the nuclear spin susceptibility, ω is the frequency, and $\omega_H = \gamma_N |AM_0|$ is the nuclear Larmor precession frequency in the hyperfine field $H_{hyp} = \pm AM_0$ of the electronic spins acting on the nuclear spins. If one introduces losses by allowing the frequency ω to become complex, the real and imaginary parts of c_{44}^* can be associated with elastic dispersion and absorption, respectively. The absorption coefficient derived in this way can be compared to Silverstein's result.[1] Note that the present result [Eq. (7)] is valid at all temperatures and includes time correlation between the spin systems.

In order to illustrate the effect of time correlation, we now outline the classical analog of Silverstein's calculation. By considering a vector model, the following effective nuclear spin-phonon interaction can be derived[9]:

$$E_{NP} = (AbM_0/K)[(I_{1x} - I_{2x})e_{xz} + (I_{1y} - I_{2y})e_{yz}] \quad . \tag{8}$$

Using this interaction to couple the elastic and nuclear spin modes and ignoring the electronic spins (except as they produce the dc hyperfine field H_{hyp}), the effective elastic constant is found to be

$$c_{44}^* = c_{44} - 2(AbM_0/K)^2 \chi_N(T)[\omega_H^2/\omega_H^2 - \omega^2] \quad . \tag{9}$$

This result is similar to Eq. (7) with the exception that the coupling to the magnon modes ($-2b^2/K$) is omitted and that the ω^2 in the numerator of the nuclear spin coupling term is replaced here by ω_H^2. The latter is the result of the exclusion of time correlation when using an effective interaction [Eq. (8)]. Clearly, near resonance (i.e., $\omega \simeq \omega_H$), this exclusion gives a negligible effect. However, in the limit $\omega \ll \omega_H$ or $\omega \gg \omega_H$, there is a marked difference; the effective interaction predicts a contribution from the nuclear spin coupling even for $\omega \to 0$ [Eq. (9)], whereas the mode coupling calculation [Eq. (7)] predicts the nuclear coupling term to go to zero as $(\omega/\omega_H)^2$.

On physical grounds, it becomes obvious that the nuclear coupling term must go to zero as $\omega \to 0$. The electronic sublattices can be treated as rigid vectors which oscillate under the influence of the magnetoelastic interaction at the frequency of the elastic strain e_{ij}. At low frequencies ($\omega \ll \omega_H$), the nuclear spins seek a configuration of minimum energy. Because of the isotropic hyperfine interaction, this configuration is collinear with electronic magnetization and therefore oscillating at the frequency ω. There exists between the two types of spin systems a phase lag ϕ determined by the frequency ω such that as $\omega \to 0$, $\phi \to 0$. Therefore, as $\omega \to 0$, the nuclei are aligned collinear with the electronic sublattices; no torques are then exerted on or by the nuclear spins. Consequently, the nuclear spin contribution to c_{44}^* must vanish as $\omega \to 0$, in agreement with Eq. (7) but not Eq. (9).

On the other hand, when using an effective interaction [e.g., Eq. (8)], the fallacious assumption is made that the effect of the hyperfine interaction on the nuclei can be separated into two independent parts: (a) a dc magnetic field acting in the equilibrium direction (taken to be the $\hat{z}$ aixs) of the electronic sublattices, and (b) a magnetic field acting in the xy plane oscillating at the frequency ω. Regardless of the frequency, the dc component of the hyperfine field exerts a torque on the nuclear spins whenever they deviate from the $\hat{z}$ axis. Therefore, even as $\omega \to 0$, the calculation gives a nuclear spin contribution to c_{44}^* [Eq. (9)]. This erroneous result is obtained by ignoring the fact that the nuclei seek a configuration collinear with the *instantaneous* position of the electronic sublattices.

Similar arguments can be used in the limit $\omega \gg \omega_H$ to demonstrate that time correlative effects are not properly taken into account when an effective interaction is used. The question of the existence of elastic dispersion at low frequencies arising from the nuclear spin system appears to be academic for the case of $\mathrm{Mn}^{55}\mathrm{F}_2$, since the effect is probably not observable. However, in a material such as RbMnF_3 with a very low magnetic anistropy, the effect should be observable at low temperatures ($T < 4$ K). (See, however, Ref. 10.)

Application of a dc magnetic field has two main effects. First, it causes a tuning of the magnon modes which can produce an increase or decrease in the strength of the magnetoelastic coupling,[6] and second, it serves to tune the nuclear spins which now experience a resultant dc field which is the vector sum of the applied and hyperfine fields. The algebra is quite tedious and we only quote the result here for $\vec{\mathrm{H}}_0 \parallel [001]$ where [001] is the axis of easy magnetization,

$$c_{44}^* = c_{44} - 2\,\frac{b^2}{K}\left(\frac{H_c^2}{H_c^2 - H_0^2}\right) - \left(\frac{AbM_0}{K}\right)^2 \left(\frac{H_c^2}{H_c^2 - H_0^2}\right) \chi_N(T)$$
$$\times\left[\left(1 - \frac{\omega_0}{\omega_H}\right)\left(\frac{\omega^2 + \omega_0\omega_H(1 - \omega_0/\omega_H)}{(\omega_0 - \omega_H)^2 - \omega^2}\right) + \left(1 + \frac{\omega_0}{\omega_H}\right)\left(\frac{\omega^2 - \omega_0\omega_H(1 + \omega_0/\omega_H)}{(\omega_0 + \omega_H)^2 - \omega^2}\right)\right] , \qquad (10)$$

where $\omega_0 = \gamma_N H_0$, $H_c \equiv (2|\lambda|K)^{1/2}$ is the spin-flop field ($H_c \simeq 93$ kG in MnF_2) and Eq. (10) is valid for $\omega \ll |\gamma_e|(H_c - H_0)$. For $H_0 \simeq 50$ kG in MnF_2 the factor $[H_c^2/(H_c^2 - H_0^2)]^2 \simeq 2$. This rather significant field dependence of the nuclear spin-phonon coupling strength contrasts with Silverstein's statement that the applied field in this configuration has no appreciable effect. Indeed, it is difficult to see how to properly include the effect of the tuning of the magnon modes in a calculation based on an effective interaction.

Although it is difficult to generalize these results to materials of different magnetic symmetry without actually carrying out the calculation for each symmetry,[10] it should be clear that in all cases the coupling strength and its temperature dependence will be determined by the factor $(AbM_0/K)^2\chi_N(T)$.[2]

In conclusion, the main motivation for the present work was to achieve a better understanding of the magnon-coupled nuclear spin-phonon interaction. The proper inclusion of both time correlation effects and magnetic field dependence has been achieved and in addition the present results are valid (to within the validity of the molecular field model) at all temperatures.

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[8] It is readily seen from Eqs. (2) and (3) or by explicit calculation that c_{44} is the only elastic constant affected by the magnetoelastic coupling and therefore also by the nuclear spin coupling. In crystals of D_{4h}^{14} tetragonal symmetry, c_{44} is obtained directly from measurements of the transverse elastic phase velocity for propagation along the [001] axis with arbitrary polarization or from propagation perpendicular to and with polarization along the [001] axis.

[9] This effective interaction results from a slight generalization of the calculation outlined in Appendix B of

Ref. 1.

[10] Note that Eqs. (7) and (10) are valid only to first order in the quantity $\eta \equiv [A^2 M_0^2 \chi_N(T)]/K$. Considering the coupling to (say) the Mn^{55} spin system at low temperatures in a material with an anomalously low magnetic anisotropy such as $RbMnF_3$ one finds that $\eta \approx 1$. The expansion is no longer valid and a more complete solution is required.

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Generalized Theory for the Temperature-Dependent Hyperfine Coupling Constant of Iron-Group *S*-State Ions*

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The orbit-lattice interaction theory describing the temperature dependence of the hyperfine coupling constant of iron-group *S*-state ions in cubic crystals is generalized by means of the same *k*-space averaging method employed in the generalization of the theory for rare-earth *S*-state ions. This generalized theory provides a good qualitative fit to experimental data on the temperature behavior of the hyperfine constant of Mn^{2+} in CaF_2, SrF_2, and BaF_2, but the calculated magnitude of the decrease in the hyperfine constant with temperature is considerably smaller (with respect to its observed value) than is the corresponding magnitude calculated from the long-wavelength theory. This disagreement between theory and experiment increases with increasing interionic distance in the host lattice. In addition, the generalized theory for iron-group *S*-state ions confirms our previous finding that optical phonons make a significant contribution to both the thermal and rigid-lattice values of the hyperfine coupling constant, and enables us to determine the range of validity of the long- and short-wavelength treatments.

In an earlier paper,[1] the orbit-lattice interaction theory for the temperature dependence of the hyperfine coupling constant of rare-earth *S*-state ions in cubic crystals was generalized by employing an expansion technique which enabled us to treat the wavelength of the lattice vibrations exactly over the entire phonon spectrum. By means of this expansion the use of the long- or the short-wavelength approximation was avoided. In this paper, we present the generalized theory for the temperature dependence of the hyperfine coupling constant of iron-group *S*-state ions in cubic crystals and apply it to the $A(T)$ data on Mn^{2+} in the alkaline earth fluorides.

It is important to note that an isotropic lattice is assumed in our generalized treatment of the *k*-space averaging, that is, we assume that each phonon branch can be decomposed into one longitudinal and two degenerate transverse modes, whereas this is true in a real lattice only at certain symmetry points in the reduced zone. Nevertheless, this is a common assumption which has been employed in orbit-lattice interaction theory since it was originally introduced,[2] and without which it would be difficult to proceed.

The expression for the orbit-lattice interaction V_{01} given in Eq. (1) of Ref. 3 can be used as a starting point in formulating the generalized theory, with the following important exception: There are three modes transforming like Γ_{5g} which vanish in the long-wavelength approximation and therefore could be ignored in Ref. 3, but they must be included in the present generalized treatment. It should also be noted that the equation derived for either the rare-earth or the iron-group ions are applicable to both the XY_6 and XY_8 molecular clusters under the relationship

$$V_{01}(XY_8) = -\tfrac{8}{9}\, V_{01}(XY_6)\ . \qquad (1)$$

The calculation of the perturbed hyperfine field for iron-group *S*-state ions proceeds in a manner analogous to the calculation for the rare-earth *S*-state ions presented in Refs. 1 and 3. The final generalized expression for the temperature-dependent hyperfine constant is[4]

$$A(T) = A(0)\{1 - D_{\mathrm{ac}} F_{\mathrm{ac}}(T) - \sum_{r}^{n-1} [D_{\mathrm{op}} F_{\mathrm{op}}(T)]_r\}, \qquad (2)$$

where the summation is taken over all optical branches of the spectrum. The constant D_{ac} is given by

$$D_{\mathrm{ac}} = \frac{8\pi}{3} \frac{g\mu_\beta \langle S_z \rangle}{H_c} \left(\frac{ee'}{R^4}\right) \sum_{n,n'} U_{nn'} \frac{81\hbar}{80\rho\pi^2 v_t^{\,3}} \left(\frac{k_B}{\hbar}\right)^2 \qquad (3)$$