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Effect of itinerant electron polarization and crystal fields upon the paramagnetism of metallic samarium compounds

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It is shown how to calculate the simultaneous effects of crystal fields, itinerant electron polarization, and interionic interactions treated in the mean-field approximation upon the paramagnetic properties of metallic samarium compounds. Expressions are derived for the paramagnetic susceptibility, the hyperfine fields, and the gyromagnetic ratio. The application of these results to the paramagnetic susceptibility of single-crystal $SmRh_4B_4$ is discussed.

INTRODUCTION

The magnetization of a metallic rare earth compound may, when the rare earth is in a well-defined valence state, be divided conceptually into the magnetization associated with the $4f$ shell and that associated with the itinerant electrons. In cases where there is no mixed valence behavior or Kondo effect these two systems are weakly coupled together, the leading term of the coupling[1] being the isotropic exchange interaction of the form $-2I(\mathbf{q})\mathbf{S}\cdot\mathbf{s}$ where $\mathbf{S}$ is the spin of the rare earth and $\mathbf{s}$ the spin of an itinerant electron. $I(\mathbf{q})$ is the strength of the interaction, $\mathbf{q}$ being the change in wave vector of the scattered itinerant electron.

This interaction has two effects. The first one, which occurs in first-order perturbation theory[2,3] is to increase the magnetic moment associated with each $4f$ shell. This occurs in two stages: first there arises the electronic analogue of the nuclear Knight shift which acts to increase the magnetic polarization of the rare earth ion if it is not already in saturation. Second, the rare earth ion induces a cloud of polarized itinerant electrons localized around it that adds to its moment. For normal tripositive rare earths the resulting change of moment is rather small, usually less than 10%, although there are exceptions such as the RCo_2 compounds in which the changes are much larger.[4] There is, in addition, the magnetization associated with the Pauli susceptibility and the Landau and core diamagnetism, lumped together as $\chi_M H$ where χ_M is the matrix susceptibility and H the applied field.

The second effect of the exchange interaction is the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling[2,5] between different ions that arises in second-order perturbation theory. This gives rise to an effective interaction of the form $-A_{ij}\mathbf{S}_i\cdot\mathbf{S}_j$ between the $4f$ spins i and j. The strength A_{ij} of the effective interaction depends on $I(\mathbf{q})$ and on the properties of the itinerant elections. This effective interaction can give rise to magnetic ordering of the $4f$ moments.

The third important influence in these materials is crystal fields. These admix and split the degenerate energy levels of the free-ion $4f$ shell by typically 100 K. In the paramagnetic state the two effects of the itinerant electrons may be treated as a perturbation upon the states of the $4f$ shell that are the eigenstates of the free-ion Hamiltonian plus the crystal field Hamiltonian.

The tripositive samarium ion has an unusual electronic structure. The ground state $J=\frac{5}{2}$ multiplet level has a Landé g factor of 2/7 and a very small paramagnetic moment of $0.845\mu_B$, resulting from antiparallel coupled $L=5$, $S=5/2$ Russell-Saunders states. The next $J=7/2$ multiplet level is only 1550 K above the ground state, the other levels are correspondingly higher. Because of this, small perturbations, such as those resulting from itinerant electron interactions, can have very large effects. Similarly, the admixture by the crystal field of more strongly magnetic excited multiplet levels into the weakly magnetic ground state can significantly influence the magnetic properties.

Calculations of the paramagnetic properties of samarium compounds have been made that take account of interionic interactions treated in the mean-field approximation and thermal occupation of higher multiplet levels but not the effects crystal fields or itinerant electron polarization and have been applied to $SmAl_3$.[6] The calculations have been extended to take account of crystal fields as well,[7-10] and it has been demonstrated that for some materials these have important consequences too. On the other hand, calculations that take account of itinerant electron polarization but not of crystal fields have shown that very large effects of polarization can occur in some cases.[3,11] It has been argued[12] that the effect of crystal fields upon the susceptibility of polycrystalline samples is less than their effect upon single crystals so these calculations may be more appropriate for the former. As examples, polycrystalline elemental samarium has a Curie constant that is almost four times smaller than the free-ion value[13] and $SmCo_2$ a Curie constant that is more than eight times larger (Ref. 4), SmZn even has a moment that is reversed in direction.[14,15] It was argued that these effects were too large to be attributed to crystal fields alone and must be due to itinerant electron polarization.

Clearly, a convincing calculation of the paramagnetic properties of metallic samarium compounds must take account of all of these effects simultaneously. The only such one that appears to have been done to date[16] is by Zhou *et al.* on $SmRh_4B_4$. Unfortunately the expression that these authors used to calculate the paramagnetic susceptibility is only approximately correct. In the present paper the exact mean-field expressions for the paramagnetic properties are derived, a discussion is given of the ionic linear response to applied fields, and comparison of theory is made with the data and fit of Zhou *et al.*[16] for $SmRh_4B_4$.

MEAN-FIELD EQUATIONS

The nature of the calculation that needs to be done is as follows. In the paramagnetic regime at a temperature T the responses of the expectation values of the operators $\langle S\rangle$ and $\langle L+2S\rangle$ of the samarium $4f$ shell to a real applied field H and to an exchange field H_{ex} are linear:

$$-\langle L+2S\rangle/\mu_B = A_{MM}(T)H + A_{MS}(T)2H_{ex} \ . \tag{1}$$

$$-\langle S\rangle/\mu_B = A_{SM}(T)H + A_{SS}(T)2H_{ex} \ . \tag{2}$$

The fields H and H_{ex} are defined by the Hamiltonian $\mathcal{H}$:

$$\mathcal{H}=\mathcal{H}_f+\mathcal{H}_x+\mu_B(L+2S)H+\mu_B S2H_{ex} \ , \tag{3}$$

where $\mathcal{H}_f$ is the Hamiltonian of the free samarium ion incorporating the central potential, Russell-Saunders coupling and spin-orbit coupling, and $\mathcal{H}_x$ is the crystal field Hamiltonian. The quantities L and S are the operators for the total orbital and spin angular momentum components of the $4f$ shell along a particular principal axis of the crystal. H and H_{ex} are the real magnetic field and the exchange field along that direction. The last two terms in $\mathcal{H}$ are small perturbations in the paramagnetic regime. The susceptibilities $A_{rs}(T)$ depend upon the electronic structure of the samarium ion, the crystal field parameters and the temperature. It will be shown in the next section that $A_{SM}(T)=A_{MS}(T)$.

The exchange field is given by $H_{ex}=-J_{ff}\langle S\rangle/\mu_B+H\alpha/2$. In this expression J_{ff} $(=\Sigma' A_{ij}$, where the prime indicates that the $j=i$ term is to be omitted) is a molecular field parameter reflecting $4f$-$4f$ coupling. This quantity comes from the RKKY interaction and contains the off-diagonal matrix elements of the $-2I(\mathbf{q})\mathbf{S}\cdot\mathbf{s}$ interaction as well as the diagonal elements.[17] The quantity α represents the effect of itinerant electron polarization upon the $4f$ shell. It is given by $\alpha=2I(0)\rho$, ρ being the Pauli susceptibility of the itinerant electrons expressed in units of density of states per atom for one spin direction. It gives rise to the electronic analogue of the nuclear Knight shift. This form of the exchange field may be derived either from the effective Hamiltonian[3] of Yosida[2] or else by calculating self-consistently each spatial Fourier component of the itinerant electron magnetization.[18]

The first step in the calculation is to substitute the expression for the exchange field into Eq. (2); this gives

$$\frac{\langle S\rangle}{-\mu_B H}=\frac{A_{SM}+\alpha A_{SS}}{1-2J_{ff}A_{SS}} \ . \tag{4}$$

Within the context of the model used here this is proportional to the transferred hyperfine field (or Knight shift) at the nucleus of a nonmagnetic neighboring atom. Equation (4) is then substituted into Eq. (1) to give

$$\frac{\langle L+2S\rangle}{-\mu_B H}=\frac{A_{MM}+\alpha A_{MS}-2J_{ff}(A_{SS}A_{MM}-A_{MS}^2)}{1-2J_{ff}A_{SS}} \ . \tag{5}$$

The other responses of the $4f$ shell may be readily obtained:

$$\frac{\langle L+S\rangle}{-\mu_B H}=\frac{A_{MM}-A_{MS}+\alpha(A_{MS}-A_{SS})-2J_{ff}(A_{SS}A_{MM}-A_{MS}^2)}{1-2J_{ff}A_{SS}} \ , \tag{6}$$

and

$$\frac{\langle L\rangle}{-\mu_B H}=\frac{A_{MM}-2A_{MS}+\alpha(A_{MS}-2A_{SS})-2J_{ff}(A_{SS}A_{MM}-A_{MS}^2)}{1-2J_{ff}A_{SS}} \ . \tag{7}$$

To obtain the total susceptibility $\chi\{=-\mu_B\langle L+2S\rangle_{\mathrm{TOTAL}}/H\}$ per $4f$ atom we need to add to Eq. (5) the itinerant electron magnetization m_e that is induced by the interaction $-2I(\mathbf{q})\mathbf{S}\cdot\mathbf{s}$; this is $m_e=-\mu_B\alpha\langle S\rangle$,[3,18] and so

$$\frac{\chi}{\mu_B^2}=\frac{A_{MM}+2\alpha A_{MS}+\alpha^2 A_{SS}-2J_{ff}(A_{SS}A_{MM}-A_{MS}^2)}{1-2J_{ff}A_{SS}}+\frac{\chi_M}{\mu_B^2} \ . \tag{8}$$

It is assumed throughout that the spectroscopic splitting factor g and the gyromagnetic ratio g' of the itinerant electrons are equal to 2. The angular momentum of the itinerant electrons is therefore[11] $\alpha\langle S\rangle/2$, and the total angular momentum $\langle L+S\rangle_T$ is obtained by adding this to Eq. (6):

$$\frac{\langle L+S\rangle_T}{-\mu_B H}=\frac{A_{MM}-A_{MS}+\frac{3}{2}\alpha A_{MS}-\alpha A_{SS}(1-\alpha/2)-2J_{ff}(A_{SS}A_{MM}-A_{MS}^2)}{1-2J_{ff}A_{SS}}+\frac{\chi_M}{2\mu_B^2} \ . \tag{9}$$

The gyromagnetic ratio g', which is proportional to the ratio of the magnetic moment of the material to its angular momentum,[11] is given by $g'=\langle L+2S\rangle_T/\langle L+S\rangle_T$, or the ratio of Eq. (8) to Eq. (9). The hyperfine field at the samarium nucleus is also given by combinations of $\langle L\rangle_T$ and $\langle S\rangle_T$.[10]

For the free-ion case when only the $J=5/2$ multiplet level is thermally occupied and there are no crystal fields $A_{MM}=a'/T+b'$, $A_{MS}=A_{SM}=(g-1)a'/gT+b'$ and $A_{SS}=(g-1)^2a'/Tg^2+b'$, where $a'=g^2J(J+1)/3k$, $b'=20/7\Delta$, g is the Landé factor of the $J=5/2$ level of the Sm^{3+} ion and Δ is the energy difference between the $J=5/2$ level and the $J=7/2$ level. For this case Eqs. (4–9) reduce to the results obtained previously.[3,11]

Dipole-dipole interactions may be incorporated into these expressions. If the field H is replaced by the sum of the applied field H_A plus a dipole field $H\rightarrow(H_A+\lambda'M)$. Then from $\chi=M/(H_A+\lambda'M)$ it follows that $H_A/H=1-\lambda'\chi$. Hence all the expressions in Eqs. (4–9) may be extended to allow for dipole fields by dividing them by the factor $(1-\lambda'\chi)$. Usually this factor is close to unity.

ISOTHERMAL LINEAR RESPONSE

The thermal average of an operator O_r is given by

$$\langle O_r\rangle_F=\frac{1}{Z}\sum_i e^{-\beta E_i'}\langle i'|O_r|i'\rangle\ , \tag{10}$$

where $\beta=1/kT$, the $|i'\rangle$ and E_i' are eigenstates and eigenvalues of the total Hamiltonian and Z is the partition function: $Z=\sum_i e^{-\beta E_i'}$. We assume that the system has been subjected to a small perturbation $-FO_s$ which consists of a field F coupled to an operator O_s; the subscript F in Eq. (10) refers to the field. As a result of this perturbation the eigenvalues E_i have become $E_i'=E_i-F\langle i|O_s|i\rangle$ and the eigenstates $|i\rangle$ have become

$$|i'\rangle=|i\rangle+F{\sum_j}'\frac{|j\rangle\langle j|O_s|i\rangle}{E_j-E_i}\ , \tag{11}$$

where the prime on the sum indicates that the $j=i$ terms are to be omitted. The terms in $\langle O_r\rangle_F$ that are linear in F arise from three sources: the change of the partition function Z due to the implicit change in the eigenvalues, the change of the eigenvalues explicitly shown in Eq. (10) and the change of the eigenfunctions (11).

The partition function is therefore

$$Z=\sum_i e^{-\beta(E_i-F\langle i|O_s|i\rangle)}\ , \tag{12}$$

and expanding the exponential in powers of F:

$$Z=Z_0(1+F\beta\langle O_s\rangle_0)+O[F^2]\ , \tag{13}$$

where Z_0 is the unperturbed partition function and $\langle O_s\rangle_0$ is the thermal average of O_s in the unperturbed state. Substituting Eq. (13) into Eq. (10) the leading terms obtained are

$$\langle O_r\rangle_F^A=\langle O_r\rangle_0-F\beta\langle O_r\rangle_0\langle O_s\rangle_0\ . \tag{14}$$

In the unpolarized state both these terms will be zero.

The second contribution to the linear response comes from substituting the change of eigenvalue into Eq. (10) but keeping the partition function and the eigenstates in their unperturbed condition. By expanding the exponential the next linear term is found to be

$$\langle O_r\rangle_F^B=\frac{F\beta}{Z_0}\sum_i e^{-\beta E_i}\langle i|O_r|i\rangle\langle i|O_s|i\rangle\ . \tag{15}$$

The final term linear in F is obtained by keeping the partition function and the eigenvalues in their unperturbed state and substituting Eq. (11) into Eq. (10). This gives

$$\langle O_r\rangle_F^C=\frac{F}{Z_0}\sum_i e^{-\beta E_i}{\sum_j}'\frac{\langle i|O_s|j\rangle\langle j|O_r|i\rangle+\langle i|O_r|j\rangle\langle j|O_s|i\rangle}{E_j-E_i}\ . \tag{16}$$

The response to order F is given by the sum of Eqs. (14)–(16).

The form of Eq. (16) is satisfactory when the states i and j are nondegenerate, but degenerate states need special treatment to remove the divergent denominator. For these states Eq. (16) is divided into halves and in one of them the dummy indices i and j are interchanged. This gives rise to a factor $(e^{-\beta E_i}-e^{-\beta E_j})$ which, when expanded and divided by the denominator of Eq. (16), gives $\beta e^{-\beta E_i}$. Then by noting that

$$\sum_i{\sum_j}'={\sum_i}'\sum_j=\sum_i\sum_j(1-\delta_{i,j})$$

if follows that those terms of $\langle O_r\rangle_T^C$ with $E_i=E_j$ may be expressed as

$$\langle O_r\rangle_F^D=\frac{F\beta}{2Z_0}\sum_i e^{-\beta E_i}\sum_j^{E_j=E_i}\langle i|O_s|j\rangle\langle j|O_r|i\rangle+\langle i|O_r|j\rangle\langle j|O_s|i\rangle-\frac{F\beta}{Z_0}\sum_i e^{-\beta E_i}\langle i|O_s|i\rangle\langle i|O_r|i\rangle\ . \tag{17}$$

The second term, which arises from the Kronecker delta, cancels Eq. (15) so the final result is

$$\langle O_r\rangle_F=\langle O_r\rangle_0-F\beta\langle O_r\rangle_0\langle O_s\rangle_0+F\chi_{rs}\ , \tag{18}$$

where

$$\chi_{rs}=\frac{\beta}{2Z_0}\sum_i e^{-\beta E_i}\sum_j^{E_j=E_i}\langle i|O_s|j\rangle\langle j|O_r|i\rangle+\langle i|O_r|j\rangle\langle j|O_s|i\rangle$$
$$+\frac{1}{Z_0}\sum_i e^{-\beta E_i}\sum_j^{E_j\neq E_i}\frac{\langle i|O_s|j\rangle\langle j|O_r|i\rangle+\langle i|O_r|j\rangle\langle j|O_s|i\rangle}{E_j-E_i}\ . \tag{19}$$

The response matrix χ_{rs} consists of two pairs of terms, one pair associated with a temperature denominator, the Curie part, and the other associated with an energy denominator, the Van Vleck part. χ_{rs} is explicitly symmetric in r,s and since each member of the pair is the complex conjugate of its partner χ_{rs} is real. For the diagonal elements χ_{rr} the four terms are real and contract to two and χ_{rr} is greater than or equal to zero. For cases where the operator O_r is L^{α} and O_s is S^{α}, α being x, y, or z, the four terms also contract to two because the two operators, being the same component of an axial vector, transform in the same way under the operations of the appropriate point group. According to the Wigner-Eckart theorem their matrix elements between basis states of irreducible representations of the group will be proportional to each other. However, the elements of χ_{rs} that are off diagonal in direction, and which will be nonzero for crystals of low symmetry whose principal axes may not be known *a priori,* will need to incorporate all four terms of Eq. (19).

It is to be noted that the Curie terms contain off-diagonal matrix elements of the operators as well as the diagonal elements. de Wijn *et al.*[9,10] omitted the off-diagonal elements in their calculation of the susceptibility along the $z[0,0,1]$ direction of cubic $SmAl_2$, but this does not affect their results because it is readily verified from group tables[19] that the matrix of an operator transforming as the z component of an axial vector is diagonal between the states of the three irreducible representations of the cubic double group. It would be necessary to take account of the off-diagonal elements to calculate the susceptibility along the x direction, for instance, if the crystal field states were quantized along the z direction, but for cubic crystals the susceptibility is isotropic so this calculation is not needed in this particular case.

The results of this section show that the response coefficients $A_{MM}=\chi_{L+2S,L+2S}$, $A_{SS}=\chi_{S,S}$, $A_{MS}=\chi_{L+2S,S}$, and $A_{SM}=\chi_{S,L+2S}$. Consequently A_{MS} is equal to A_{SM}.

SUSCEPTIBILITY OF $SmRh_4B_4$

The susceptibility of polycrystalline $SmRh_4B_4$ was first measured by Hamaker *et al.*[20] who found that its susceptibility could be fitted to a Curie-Weiss law $\chi=\chi_M+\mu_B^2p_{\text{eff}}^2/3k(T-\theta)$. From the effective paramagnetic moment p_{eff}:

$$p_{\text{eff}}=p_0[1+\alpha(g-1)/g-(1+\alpha)\theta/T_0]\ , \tag{20}$$

where θ is the paramagnetic Curie temperature and $T_0=322$ K, the value of $\alpha=+0.022$ was deduced.[13] It was argued[13] that this demonstrated that itinerant electron effects were unusually small in this compound. Equation (20) is obtained from Eq. (8) when the free-ion values of the response coefficients are used.[3,11,22]

The susceptibility of single crystal $SmRh_4B_4$ was measured by Zhou *et al.*[16] They fitted their results with a theory which took account of crystal field effects, interactions and itinerant electron polarization. They used the crystal field parameters previously deduced for the rare earth-Rh_4B_4 series of compounds.[22] These gave a doublet ground state that was *roughly* $|5/2,+/-1/2\rangle$, with a $|5/2,+/-3/2\rangle$ doublet at 22 K above that and another $|5/2,+/-5/2\rangle$ doublet at 190 K. Even though in this material itinerant electron effects are unusually small[13] it was found it was essential to include them to obtain a satisfactory fit to the susceptibility. The expression that Zhou *et al.* used was[16]

$$\chi=\chi_0\frac{\left[1+\beta\dfrac{\langle S\rangle}{\langle L+2S\rangle}\right]}{1-\gamma\chi_0}\ , \tag{21}$$

where χ_0 is the crystal field only susceptibility and $\beta=+0.064$. The above equation does not agree with Eq. (8) derived in this paper, but an examination of it shows that in this case it may not be too bad an approximation. First of all the matrix susceptibility is negligible.[16] Second, the ordering temperature is so low that the term $\gamma\chi_0$ that represents interionic interactions is generally very much less than unity. Lastly, for small α we are comparing $2\alpha A_{MS}/A_{MM}$ with $\beta\langle S\rangle/\langle L+2S\rangle$. The leading terms of the latter two factors are both $(g-1)/g$ so we identify $\alpha=\beta/2=+0.032$. This value of α has the same sign and magnitude as that of $+0.022$ obtained from the polycrystalline susceptibility. Assuming α to be $+0.032$ and taking[16] $\rho=0.35$ eV^{-1}, a value of $+0.0457$ eV is obtained for $I(\mathbf{O})$. This value gives $I(\mathbf{O})^2\rho=7.3\times10^{-4}$ eV. This compares with a value of $I^2\rho=3.4\times10^{-4}$ eV for the value of $I(\mathbf{q})$ averaged over the Fermi surface obtained from an analysis of superconducting transition temperatures of the other $R\text{Rh}_4\text{B}_4$ compounds in the series.[23]

The comparison is reasonable considering that the $I(\mathbf{q})$ are small quantities. Although a full analysis of the susceptibility of $SmRh_4B_4$ using Eq. (8) remains to be carried out, the approximate analysis discussed here illustrates the importance of accounting for itinerant electron polarization effects in metallic samarium compounds even in cases where they are small. It demonstrates too that the value of α obtained from a Curie-Weiss fit to the polycrystalline susceptibility[3,11] gives a useful estimate of the true size of this quantity obtained by fitting single crystal data.

Note added in proof. For the case of $r=s$, it can be shown that the response in Eq. (18) that is linear in field is positive definite by the method of L. D. Landau and E. M. Lifshitz, in *Statistical Physics,* Part 1, 3rd ed. (Pergamon, Oxford, 1987), Sec. 32.

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