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Conduction-electron polarization in intermetallic actinide compounds

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The large conduction-electron polarization observed in many intermetallic actinide compounds with well localized $5f$ moments is analyzed using a phenomenological model for the electron density of states. The model is applied to PuP and successfully predicts appreciable polarization effects existing in the paramagnetic phase. Possible consequences of this model in our understanding of the electronic structure of other actinide compounds with the NaCl-type structure are discussed.

I. INTRODUCTION

Experimental investigation of the magnetic properties of metallic actinide systems has established that the electronic structure of the actinide ions is considerably more complicated than that of the rare-earth ions.[1] The rare-earth ions, even in metallic systems, adopt ionic ground configurations, and the $4f$ electrons are well localized. The magnetic properties of rare-earth intermetallic compounds can be interpreted satisfactorily with a localized-$4f$-electron model. On the other hand, the $5f$-electron wave functions in actinide elements are spatially more extended than the corresponding $4f$ wave function in the rare earths. The large extension of the $5f$ wave function is responsible for the large modification of the atomic $5f$ electron states by the crystalline electric field, by $5f$-$5f$ and $5f$-$6d$ overlap between two actinide sites, and by hybridization with the s and p electrons of the ligands. The sensitivity of the $5f$ electrons to their crystalline environment results in a variety of magnetic behaviors which may not be easily interpreted with a single-band or localized-$5f$-electron model.[2-6] The effects of strong angular anisotropy of the $5f$-electron orbitals on the exchange interactions and on the magnetic properties of NaCl-type compounds have been discussed by Chan[7] and later substantiated by neutron-scattering experiments.[8] The purpose of the present paper is to focus attention on the polarization of the conduction electron by the $5f$ electrons both in the ordered magnetic phase and in the paramagnetic phase in NaCl-type compounds.

The direct evidence of large conduction-electron polarization is shown by the difference in the value of the magnetic moment in the ordered state as measured by magnetization and neutron scattering experiments. The magnetization measurement gives the net total magnetic moment whereas the neutron measurement performed at small scattering angle and extrapolated to zero angle gives the localized magnetic moment. The difference in these two measured magnetic moments, $\Delta\mu = \mu_{\text{tot}} - \mu_{\text{loc}}$, can provide a measure of the conduction-electron spin polarization (CEP). This difference is generally negative and can be as large as $-0.4\mu_B$. This negative moment implies that the majority spin of the conduction electrons is antiparallel to the local moment. The large negative conduction-electron polarization exists in several actinide compounds as well as in the interstitial regions of transition-metal ferromagnets. In PuP, a comparison between neutron-diffraction and magnetization experiment yields $\Delta\mu = (-0.35 \pm 0.07)\mu_B$.[9]

For an understanding of this large polarization effect in actinide compounds, we study the electronic structure of PuP in terms of a model calculation. We have chosen this system for our investigation since there exists a large body of experimental data on Knight shift, nuclear spin-lattice relaxation time, magnetization, and neutron-scattering form factors. The analysis presented here can be applied to any other actinide compounds.

II. EMPIRICAL DETERMINATION OF DENSITY OF STATES

In order to understand the conduction-electron polarization in both ordered and paramagnetic phases of PuP, we need some information about its electronic structure. Unfortunately, no band-structure results are yet available on this compound. However, it is possible to obtain qualitative features about the band structure for PuP from the calculations of Davis[10] on uranium compounds with NaCl structure. The conduction band consists of primarily d and s electrons. The d band is split into t_{2g} and e_g with the t_{2g} band lying below the Fermi energy E_F. The s band lies higher than the t_{2g} band and is, in general, close to the Fermi energy.

We assume that the density of states is composed of an s-like free-electron band and a quasilocalized 6*d* state. The 5*f* electrons, as mentioned earlier, are localized well below the conduction-band continuum and are not expected to contribute to the observed conduction-electron polarization.

To calculate the density of states of *s* and *d* electrons at the Fermi energy separately we make use of the results of nuclear magnetic resonance (NMR) and magnetic susceptibility experiments. The exchange coupling parameter, J_{cf} between the localized 5*f* electrons and conduction electrons can also be calculated on the basis of the uniform polarization model.

The magnetic susceptibility χ and the Knight shift of the ligand nucleus K in intermetallic actinide compounds can be given by

$$\begin{aligned} K &= K_0 + K_f , \\ \chi &= \chi_0 + \chi_f , \end{aligned} \tag{1}$$

where K_0 and χ_0 represent contributions from *s* electrons and are approximately temperature independent. K_f and χ_f, on the other hand, are generally temperature dependent and are thought to arise from localized 5*f* electrons. In the indirect exchange coupling model, K_f can be expressed by the relation[11]

$$\frac{dK_f}{d\chi} = J_{sf}(0) N_s(E_f) H_{\text{hfs}} \frac{d\langle S_z\rangle / H}{d\chi} , \tag{2}$$

where $N_s(E_f)$ is the *s*-electron density of states for one spin direction at the Fermi level and H_{hfs} is the contact hyperfine field. $J_{sf}(0)$ is the exchange coupling parameter per spin determined by the Fourier component of the effective *s-f* exchange integral for momentum transfer $\vec{q} = \vec{K} - \vec{K}' = 0$.

The expression for the spin projection and susceptibility of the localized f^n configuration is[3]

$$\frac{\langle S_z\rangle}{H} = \frac{\mu_B}{Z} \left(\frac{1}{kT} \sum_{\Gamma} e^{-E_\Gamma/kT} \sum_{\Gamma'}{}' \langle\Gamma|\mu_z|\Gamma'\rangle\langle\Gamma'|S_z|\Gamma\rangle + \sum_{\Gamma} \sum_{\Gamma'}{}'' \langle\Gamma|\mu_z|\Gamma'\rangle\langle\Gamma'|S_z|\Gamma\rangle \times \frac{e^{-E_\Gamma/kT} - e^{-E_{\Gamma'}/kT}}{E_{\Gamma'} - E_\Gamma} \right) \tag{3}$$

and

$$\chi_{\text{ion}} = \frac{\mu_B^2}{Z} \left(\frac{1}{kT} \sum_{\Gamma} e^{-E_\Gamma/kT} \sum_{\Gamma'}{}' |\langle\Gamma|\mu_z|\Gamma'\rangle|^2 + \sum_{\Gamma} \sum_{\Gamma'}{}'' |\langle\Gamma|\mu_z|\Gamma'\rangle|^2 \frac{e^{-E_\Gamma/kT} - e^{-E_{\Gamma'}/kT}}{E_{\Gamma'} - E_\Gamma} \right) \tag{4}$$

where $\sum'$ sums over states Γ' that are identical to or degenerate with $|\Gamma\rangle$, and $\sum''$ sums over states $|\Gamma'\rangle$ that are distinct from and nondegenerate with $|\Gamma\rangle$. Z is the partition function.

A detailed analysis of the neutron elastic magnetic cross section of PuP measured at 4.2 K, including the effects of intermediate coupling and J mixing, allows an identification of the electronic ground state of the Pu ion.[9] In fitting the experiment and nonperturbative crystal-field theory quantitatively, the sign and the approximate magnitude of the two cubic-crystal-field parameters were determined. The values for the two crystal-field parameters are $A_4\langle r^4\rangle \approx -450$ cm^{-1} and $|A_6 \langle r^6\rangle| \leq 30$ cm^{-1}. With this crystal-field strength and in the absence of the exchange field, the Γ_7 and Γ_8 levels are quite closely spaced (~15 cm^{-1}) with Γ_7 lower in energy. In the present analysis, we use $A_4\langle r^4\rangle = -450$ cm^{-1} and $A_6\langle r^6\rangle = -5$ cm^{-1} to calculate the eigenvectors and eigenvalues of the Pu^{3+} ion in PuP. From the energy-level diagram, we obtain

$$\frac{d\langle S_z\rangle / H}{d\chi} = -1.41 . \tag{5}$$

Combining this result with the experimental value[12] for $dK/d\chi = 16.7$ (emu/mole)$^{-1}$, and taking $H_{\text{hfs}} = 760$ kOe, we find from Eq. (2)

$$J_{sf}(0) N_s(E_F) = -8.697 \times 10^{-2} . \tag{6}$$

To calculate the *s-f* exchange coupling parameter $J_{sf}(0)$ and the *s* like density of states $N_s(E_F)$ separately, we make use of the measured spin-lattice relaxation time, $T_{1f}T$ at the phosphorous site due to the coupling with the localized 5*f* electrons. This is given by[11]

$$\frac{d(T_{1f}T)}{dT} = [4\pi\gamma_n^2 \hbar k H_{\text{hfs}}^2 N_s^3(E_F) |J_{sf}(0)|^2]^{-1} \times \frac{d}{dT} [G_0^{zz}(0)]^{-1} . \tag{7}$$

In the crystal-field model, the expression for the spectral density G_0^{zz} for the localized f^n configuration is

$$G_0^{zz}(0) = \frac{2}{z} \left(\frac{1}{kT} \sum_{\Gamma} e^{-E_\Gamma/kT} \sum_{\Gamma'}{}' |\langle\Gamma|S_z|\Gamma'\rangle|^2 + \sum_{\Gamma} \sum_{\Gamma'}{}' |\langle\Gamma|S_z|\Gamma'\rangle|^2 \frac{e^{-E_\Gamma/kT} - e^{-E_{\Gamma'}/kT}}{E_{\Gamma'} - E_\Gamma} \right). \tag{8}$$

For the localized spins in the paramagnetic phase, we obtain

$$\frac{d}{dT} [G_0^{zz}(0)]^{-1} = 4.241 \times 10^{-5} \text{ eV/°K} . \tag{9}$$

Substituting Eq. (9) and the experimental value[11] of $d(T_{1f}T)/dT = 2.3 \times 10^{-4}$ sec^{-1} in Eq. (8), we obtain

$$J_{sf}^2(0) N_s^3(E_F) = 9.262 \times 10^{-3} \text{ eV}^{-1} . \tag{10}$$

Equations (6) and (10) can now be combined to yield

$$J_{sf}(0) = -7.1 \times 10^{-2}\ \text{eV/spin}$$

and (11)

$$N_s(E_F) = 1.22\ \text{states/(eV atom spin)}.$$

The apparent large value of $N_s(E_F)$ is due to the overlap of the f electron of the actinide ion with the s electron of the phosphorous. The physical interpretation of $N_s(E_F)$ in terms of effective mass is given in Sec. III.

The density of states of d electrons at the Fermi energy $N_d(E_F)$ can be obtained using the uniform polarization model in the ferromagnetic region,

$$\Delta\mu = J_{cf}(0)N(E_F)\langle S_z\rangle = 0.39\mu_B. \tag{12}$$

Here $N(E_F)$ is the total density of states for electrons of both spins at the Fermi energy. The value of $\Delta\mu$ is obtained by subtracting the saturation value of $0.42\mu_B$ from the calculated value of $\mu_Z = 0.81\mu_B$ using the crystal-field parameters specified earlier. Using calculated values of $\langle S_z\rangle = -1.543$ at 5 K in the ordered state, we obtain

$$J_{cf}(0)N(E_F) = -0.253. \tag{13}$$

From Eq. (11) and the sign of $\langle S_z\rangle$, it is clear that the conduction electrons are ferromagnetically coupled to $\langle S_z\rangle$ and antiferromagnetically coupled to μ_z. Since both s and d electrons lie fairly close to the Fermi energy, we assume that

$$J_{sf}(0) = J_{df}(0) = J_{cf}(0). \tag{14}$$

The total density of states $N(E_F)$ can now be obtained from Eqs. (11), (13), and (14),

$$N(E_F) = 3.56\ \text{states/(eV atom)}. \tag{15}$$

Using Eqs. (11) and (15) we obtain the following result for the density of 6d electrons per spin at the Fermi energy,

$$N_d(E_F) = 0.55\ \text{states/(eV atom spin)}. \tag{16}$$

In calculating $N_d(E_F)$, we have made the implicit assumption that the total density of states at the Fermi energy is composed of s- and d-like electrons.

III. MODEL DENSITY OF STATES AND CONDUCTION-ELECTRON POLARIZATION

In order to calculate the contribution to the total moment due to conduction-electron polarization, we use the molecular field theory and a model density of states. The total Hamiltonian for the localized and conduction electrons is given by

$$\begin{aligned} H = H_c + \sum_{k\sigma} \epsilon_k a^\dagger_{k\sigma} a_{k\sigma} - \frac{1}{2}\sum_{i,j} J_{ij}\vec{S}_i\cdot\vec{S}_j - \frac{1}{N}\sum_i \sum_{k,k'} J_{cf}(\vec{k},\vec{k}')e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_i}\,[S^z_i(a^\dagger_{k\uparrow}a_{k'\uparrow} - a^\dagger_{k\downarrow}a_{k'\downarrow}) + S^+_i a^\dagger_{k\downarrow}a_{k'\uparrow} + S^-_i a^\dagger_{k\uparrow}a_{k'\downarrow}] \\ -\frac{\mu_B H}{2N}\sum_i \sum_{k,k'} e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_i}(a^\dagger_{k\uparrow}a_{k'\uparrow} - a^\dagger_{k\downarrow}a_{k'\downarrow}) - g\mu_B H\sum_i (L^z_i + 2S^z_i), \end{aligned} \tag{17}$$

where H_c is the crystal-field Hamiltonian. The third term is the exchange interaction between the localized spins arising from sources other than the conduction electrons. The fourth term describing the conduction-electron polarization also contributes to the exchange term through a second-order process. The last two terms incorporate the effect of an external magnetic field applied along the z direction. It is to be noted here that we neglect the orbital polarization in the first approximation. The Green's function of the conduction electrons in the molecular field approximation can be obtained as

$$\langle\langle a_{k\pm} ; a^\dagger_{k'\pm}\rangle\rangle = \frac{(1/2\pi)\delta_{kk'}}{E - E_k \pm \mu_B H \pm J_{cf}(0)\langle S_z\rangle}, \tag{18}$$

where +(−) corresponds to the conduction electron with spin ↑(↓).

Using the above equation and the following model for the energy dependence of the density of states, the polarization of the conduction electrons can be calculated in a straightforward manner. Since the s electrons are relatively free and d electrons fairly localized, we express the dispersion of s like density of states $N_s(E)$ by a parabola and the d density of states $N_d(E)$ by a truncated Lorentzian i.e.,

$$N_s(E) = (1/2\pi^2)(2m^*/\hbar^2)^{3/2}E^{1/2}\Omega_0 \tag{19}$$

and

$$N_d(E) \begin{cases} = \dfrac{1}{2\pi^2}\left(\dfrac{2m^*}{\hbar^2}\right)^{3/2} \dfrac{E_F^{5/2}\Omega_0}{(E-E_0)^2+\Delta^2} & \text{for } E_0 - \Delta \leqslant E \leqslant E_0 + \Delta \\ = 0, & \text{otherwise.} \end{cases} \tag{20}$$

Here m^* is an adjustable parameter which has the significance of the effective mass m^*/m of the conduction electrons. Ω_0 is the volume per atom which for PuP equals $152.2a_0^3$. The peak in the d density of states occurs at energy E_0 and Δ is a measure of the width of the resonant d states. The partial density of states in Eqs. (19) and (20) are shown schematically in Fig. 1 for illustration.

Our model density of states contains four parameters, namely, m^*/m, E_F, E_0, and Δ. Out of these, three parameters can be fixed independently by using the empirical values of s and d density of states at the Fermi energy as given by Eqs. (11) and (16) and by noting that the total density of states $N(E) = N_s(E) + N_d(E)$ must integrate to give four states per atom (Pu is in the 3+ and P is in the 5+ state), i.e.,

$$\int_0^{E_F} N_s(E)\,dE + \int_0^{E_F} N_d(E)dE = 4 \text{ states/atom}. \tag{21}$$

Since our subsequent calculations show that the value of conduction-electron polarization is insensitive to the choice of the Fermi energy, we have taken $E_F = 2$ eV for the remaining calculation. Using this value for E_F and Eq. (11) we integrate the s density of states in Eq. (19) and find $m^*/m = 5.01$ and 3.25 states/atom having s character. This result combined with Eq. (21) yields 0.75 d states/atom. Thus, using Eq. (20), we have

$$\begin{aligned} 0.75 &= \int_0^{E_F} N_d(E)\,dE \\ &= \frac{1}{2\pi^2}\left(\frac{2m^*}{\hbar^2}\right)^{3/2} \frac{E_F^{5/2}\Omega_0}{\Delta} \\ &\quad \times \{\tan^{-1}[(E_F - E_0)/\Delta] - \tan^{-1}(-1)\}. \end{aligned} \tag{22}$$

Solving the nonlinear Eqs. (22), (20), and (16), we obtain $\Delta = 2.45$ eV and $E_0 = 3.67$ eV. It should be noted here that if we take an alternate model of the density of states with d electrons lying below the tail of the s band, i.e., E_0 to be negative, no reasonable value of the parameters could be found.

Using Eqs. (17), (18), and (19), we obtain the following contributions to the moment due to the polarization of s and d conduction electrons.

$$\begin{aligned} M_s = \frac{1}{3}\frac{C_d}{E_F^{5/2}} \{ &[E_F + \mu_B H + J_{cf}(0)\langle S_z\rangle]^{3/2} \\ &- [E_F - \mu_B H - J_{cf}(0)\langle S_z\rangle]^{3/2}\}\mu_B \end{aligned} \tag{23}$$

and

$$\begin{aligned} M_d = \frac{C_d}{2\Delta}\bigg(&\tan^{-1}\frac{E_F + \mu_B H + J_{cf}(0)\langle S_z\rangle - E_0}{\Delta} \\ &- \tan^{-1}\frac{E_F - \mu_B H - J_{cf}(0)\langle S_z\rangle - E_0}{\Delta}\bigg)\mu_B, \end{aligned} \tag{24}$$

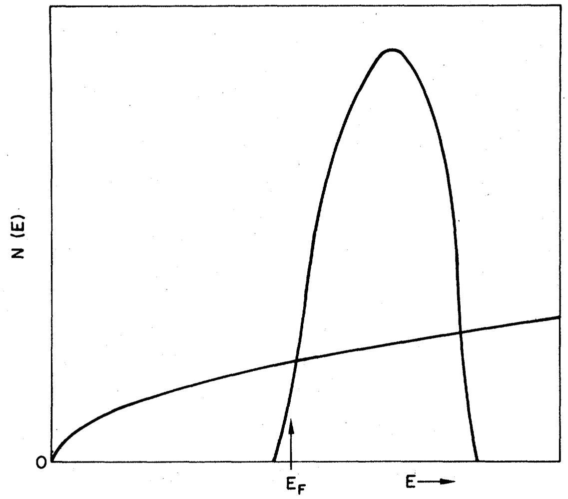

FIG. 1. Schematic electron density of states for PuP. The sp-like band is assumed to be parabolic, whereas the d-band has the shape of a truncated Lorentzian.

with

$$C_d \equiv (1/2\pi^2)(2m^*/\hbar^2)E_F^{5/2}\Omega_0.$$

In the ordered phase in the absence of an external field, we obtain

$$M_d = 0.12\mu_B \tag{25}$$

and

$$M_s = 0.27\mu_B. \tag{26}$$

Thus the total contribution to the moment from the conduction-electron polarization is $0.39\mu_B$ and the moment is *antiparallel* to the localized moment derived from neutron scattering experiments. This agrees very well with the experimental value of $(-0.35 \pm 0.07)\mu_B$. Since the uniform-polarization model used in Eq. (12) is essentially the same as the molecular field approximation we get the same total contribution to the moment from the conduction-electron polarization as used in Eq. (12). This good agreement should be treated with caution not only because we have used a model density of states but because we have also neglected contributions from orbital effects. Nevertheless, the fact that this simple model based on a molecular field approximation is capable of yielding a large negative conduction-electron polarization is encouraging. Furthermore, the value of the polarized moment is not sensitive to the details of the band structure since the uniform-polarization model does not take into account the fine structure in the density of state.

We now use this band model to calculate the magnetic susceptibility due to the conduction

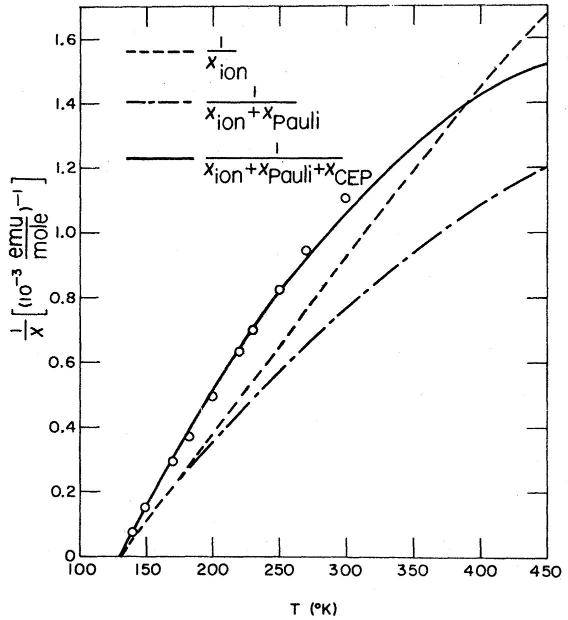

FIG. 2. Comparison of calculated and experimental reciprocal susceptibility ($1/\chi$) as a function of temperature. The open circles are experimental values taken from Ref. 12.

electrons in the paramagnetic phase. In this phase a $5f$-electron spin moment $\langle S_z\rangle$ is induced by the external field H and this is likely to cause polarization of the conduction bands. We, therefore, expect to get a term proportional to $\langle S_z\rangle$ in the total susceptibility χ. Using expressions for M_s and M_d as given in Eqs. (23) and (24), and making the assumption that $\mu_B H \ll \Delta$, we get the following contributions to χ in the paramagnetic phase arising from the conduction electrons, as well as local moments,

$$\chi_c = \chi_{\text{Pauli}} + \chi_{\text{CEP}} + \chi_{\text{ion}}\,, \tag{27}$$

where

$$\chi_{\text{Pauli}} = N(E_F)\mu_B^2 \tag{28}$$

and

$$\chi_{\text{CEP}} = -N(E_F)J_{cf}(\langle S_z\rangle/H)\mu_B\,. \tag{29}$$

Using the parameters described earlier, χ_{Pauli} and χ_{CEP} can be evaluated as a function of temperature. Ignoring the effect of temperature on the band density of states at the Fermi energy, we find that χ_{Pauli} is temperature independent while χ_{CEP} depends on temperature through $\langle S_z\rangle/H$. $\chi_{\text{ion}}(T)$ in the paramagnetic phase of PuP is calculated as a function of temperature using the eigenvector and eigenvalues of the Pu^{3+} ion derived from the neutron scattering experiment mentioned earlier. For comparison with experiment in the paramagnetic phase, we have shifted the zero of the inverse of $\chi_{\text{ion}}(T)$ to the experimental magnetic ordering temperature. The various contributions to susceptibility are compared with the available experimental data in Fig. 2. It is clear that the conduction-electron polarization contribution to paramagnetic susceptibility is large and essential for a quantitative understanding of the experimental data.

We have thus been able to develop a model of the density of states for PuP which can consistently explain the conduction-electron contribution to the ordered moment in the ferromagnetic phase and to the susceptibility, χ in the paramagnetic phase and is consistent with the ^{31}P Knight shift and T_1T. One can make the following conclusions about the electronic structure of PuP on the basis of this model density of states.

(i) The conduction-electron polarization in PuP is large due to the fact that the coupling parameter $J_{cf}(0)$ is fairly high. The d band is fairly narrow and contributes less than the s electron to the conduction-electron polarization. The same picture might hold good in other actinide compounds where fairly large conduction-electron polarization effects are observed.

(ii) There are expected to be many consequences of a narrow band with E_F lying at a critical position of the density of states. Shifting of the Fermi energy E_F or altering the width of the d band by applied pressure or doping with another element may produce drastic effects on the conduction-electron polarization. The structural transition associated with the magnetic transitions in NpX compounds[13] may be due to the occurence of a narrow d band (or more correctly t_{2g} band). Particularly the occurrence of a tetragonal phase in NpAs around 142 K is possibly associated with the effect of conduction-electron polarization on the density of states with change of magnetic structure.

(iii) The relative contribution of the s electrons to the CEP is fairly high and consequently any calculation of the polarization effect should include both s and d electrons. This is in contrast to the case of rare-earth intermetallic compounds where the contribution from s electrons is relatively small.

(iv) Both s and d bands are close to the Fermi energy E_F with the tail of the s band lying below the peak of the t_{2g} band. The existence of a narrow t_{2g} band near E_F is essential not only for explaining a significant portion of the conduction-electron polarization, but also for accounting for the sign and magnitude of the crystal-field parameter $A_4\langle r^4\rangle$.[14] In addition, consistent explanation of all experimental results for PuP is only possible if the *s* band lies below the *d* band. In this regard, the band structure calculated by Davis[10] for NaCl-type actinide compounds would not be able to explain the experimental results in PuP. The calculation of the conduction-electron polarization presented here rests on the molecular-field-type decoupling of the localized and conduction-electron spins. For the case of fairly large interaction between localized and conduction-electron spins, it is necessary to go beyond molecular field approximation. Such a calculation is presently under investigation.

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