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Nonlinear modification of the hybridization interaction due to band polarization in the magnetically ordered state in f^1 systems

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The hybridization between the f level and the band electrons changes the properties of the band states. In a magnetically ordered system, where the ordering results from the hybridization-mediated two-ion interaction, as in the cerium monopnictides, the band density of states at the Fermi level can be changed. This, in turn, can change the two-ion interaction which is mediated by the band, as in the Ruderman-Kittel-Kasuya-Yosida interaction. Such a change depends on the type of magnetic structure. Under certain conditions, this effect can increase the anisotropy favoring moment alignment along the cube-edge direction.

I. INTRODUCTION

The mixing between moderately delocalized f levels and the band electrons in cerium and light actinide materials is a key factor in determining the magnetic properties of some compounds. As discussed elsewhere,[1,2] in cerium and light actinide monopnictides, the dominant interionic coupling arises from the resulting hybridization-mediated interaction, which is transmitted by the band electrons as in the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction.[3] The resulting two-ion interaction is highly anisotropic and has been used to explain the magnetic structures obtained in the monopnictides of cerium,[1] uranium[4,5] and plutonium.[6] The interaction between the localized and the band electrons also changes the properties of the band electrons. As we shall show below, in magnetically ordered systems, the modification introduced in the band dispersion depends on the magnetic quantum number of the band electrons and this causes band polarization. The two-ion interaction, which depends on the band density of states (DOS), is modified as the band becomes polarized in response to magnetic ordering of the ions. Hence the two-ion interaction itself depends on the nature of the magnetic structure. This effect will be explored in detail. As a case of interest, the modification in the coupling introduced in the $f^1(\mathrm{Ce}^{3+})$ systems to higher orders in the mixing strength (between localized and band states), and its effect on the magnetic anisotropy in CeBi will be investigated.

We model the interaction of the $\mathrm{Ce}^{3+}(f^1)$ ion with the band electrons by the Anderson[7] model, which treats the mixing of a localized state with the band. The Hamiltonian $\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1$ is given by

$$\mathscr{H}_0=\sum_{km}\epsilon_k n_{km}+\sum E_0 n_m+U/2\sum_{m\neq m} n_m n_{m'}\ , \tag{1}$$

$$\mathscr{H}_1=\sum_{km}(V_k c_{km}^\dagger c_m+V_k^* c_m^\dagger c_{km})\ . \tag{2}$$

Here $m\ (=m_j)$ represents the z component of the total angular momentum $j\ (=\frac{5}{2})$ about the mixing site. c_m and c_{km} are the annihilation operators for the localized and the band states respectively; n_m and n_{km} are the corresponding number operators. $\mathscr{H}_0$ contains the energy of the band electrons, the energy E_0 of the localized state (which in the compounds of interest is usually below, but close to, the Fermi energy), and the Coulomb correlation energy U between f electrons in different localized states denoted by their m values. U is usually large (~ 5 eV) for magnetic systems. $\mathscr{H}_1$ describes the mixing of the localized state with the band; the mixing potential V_k is assumed to be spherically symmetric, and hence mixes only states of the same m about any ionic site. We shall show that the mixing affects the band in an important way; it changes the DOS of the band due to the exclusion of the band states in the vicinity (in energy) of the localized state. In turn, this change in the band structure affects the two-ion interaction which depends on the band DOS at the Fermi level.

In the absence of $\mathscr{H}_1$, the energy of the localized state of magnetic quantum number m is given by

$$E_m=E_0+U\sum_{m'\neq m}\langle n_{m'}\rangle\ . \tag{3}$$

When the average occupation numbers $\langle n_m\rangle$ of the different $|m\rangle$ states differ, either due to single-ion effects (e.g., external magnetic field), or due to a magnetically ordered state brought about by a two-ion interaction [such an interaction can be derived as a consequence of the Hamiltonian $\mathscr{H}$, see Eqs. (9)—(11) below], the energies E_m differ significantly. Depending on the energies E_m of the localized states, the different band states undergo changes in their dispersion at different energies, leading to a rearrangement of their populations and hence a net polarization of the bands.

In our previous theory[1] for the behavior of CeBi, the model parameters which reproduce equilibrium behavior in zero external magnetic field (paramagnet → type I antiferromagnet → type IA antiferromagnet with decreasing temperature), predict too small a value for the anisotropy field (80 kOe) that is required to change the direction of moment away from the cube-edge direction $\langle 001\rangle$, which is the easy direction of magnetization, to the body-diagonal direction $\langle 111\rangle$; experimentally, the anisotropy favoring alignment along the cube-edge direction exists for magnetic fields at least up to 120 kOe for CeBi and 150 kOe for CeSb (limit of available experimental data).[8,9] We believe that band polarization effects in the ordered ferromagnetic state (in CeBi a $\langle 001 \rangle$ ferromagnetic state is stabilized at low temperature by an external magnetic field of about 40 kOe) may be responsible for increasing the planar coupling strength and thus increasing the anisotropy favoring moment alignment along the cube-edge direction.

The two-ion interaction, which is mediated by the band, depends on the number of band states at the Fermi level as in the RKKY interaction. The DOS at the Fermi surface can be changed due to the mixing with the f level, and in the magnetically ordered state, this DOS depends on the occupation of the localized states which are denoted by their magnetic quantum number m $(\neq m_j)$. The m dependence of the DOS can, in turn, modify the strength, as well as the symmetry of the hybridization-mediated two-ion interaction. Thus a "bootstrapping" (nonlinear effect) situation exists in determining the two-ion interaction.

In the following sections, we shall discuss, starting from the Hamiltonian of Eq. (1) and (2), the origin of the band polarization and its effect on the two-ion coupling in the presence of band DOS distortion effects. The theory of this effect will be discussed for the simplest case, that of $\mathrm{Ce}^{3+}(f^1)$, for a ferromagnetically ordered system. Implications for f^n $(n>1)$ will be mentioned briefly. The qualitative nature of the theory at present precludes the possibility of calculations for the cases where the ion contains more than one f electron.

II. BAND DISTORTION

A. Origin

Starting from the Hamiltonian $\mathcal{H}$ of Eqs. (1) and (2), Anderson[7] has derived the Green's function of the band electrons as

$$G_{kk}^m(E)=\frac{1}{E-\epsilon_k}+\frac{|V_k|^2}{(E-\epsilon_k)^2(E-E_m+i\Delta_m)}\ , \tag{4}$$

where ϵ_k is the unperturbed energy of the band state of wave vector $\mathbf{k}$, and is independent of m. E_m and Δ_m are the position and the width (due to the mixing) of the localized state m; E_m is given by Eq. (3) and Δ_m is given by[7]

$$\Delta_m=\pi\langle V^2\rangle\rho(E_m)\ , \tag{5}$$

where $\rho(E)$ is the density of states (DOS) at E. It is seen from Eq. (4) that the poles of the Green function occur at exactly the same energies ϵ_k as in the unperturbed case, but the DOS has changed due to the mixing.

The DOS, which is related to the imaginary part of the Green's function, is

$$\rho^m(E)=-\frac{1}{\pi}\int d^3k\,\mathrm{Im}G_{kk}^m(E)\ . \tag{6}$$

The imaginary part of the Green function G_{kk}^m of Eq. (6) has three parts: (i) There is a first-order pole arising from $(E-\epsilon_k)^{-1}$ which reproduces the unperturbed band DOS; the second part (ii) is

$$\left[\mathrm{Re}\frac{|V_k|^2}{(E-\epsilon_k)^2}\right]\left[\mathrm{Im}\frac{1}{E-E_m+i\Delta_m}\right]\ ;$$

and the third part (iii) is

$$\left[\mathrm{Im}\frac{|V_k|^2}{(E-\epsilon_k)^2}\right]\left[\mathrm{Re}\frac{1}{E-E_m+i\Delta_m}\right]\ .$$

The contribution from (ii) is of $O(V^4)$ as Δ_m is of $O(V^2)$. Moreover this term contributes only to the mixing with the local state and is not a true polarization. Contribution (iii) is of $O(V^2)$ and is calculated from Eq. (4) using a contour integration to find the residue at the second-order pole at ϵ_k. The resulting DOS is

$$\rho^m(E)=\rho_0^m(E)+\frac{d\rho_0}{dE}\frac{|V|^2(E-E_m)}{(E-E_m)^2+\Delta_m}\ , \tag{7}$$

where ρ_0 is the unperturbed DOS.

In deriving the Eq. (7), V_k has been treated as a constant V, i.e., we assume that V_k does not change significantly with energy. Even though, in the derivation, m labels the z component of band angular momentum about a particular site, we use m to denote the band which is made up of these m components. This involves an important assumption, that there is no substantial overlap of the charge densities which produce these spherical components at each ionic site. Such a problem does not exist for the dilute system, considered by Anderson,[7] Coqblin and Schrieffer,[10] and Cornut and Coqblin.[11] This assumption is essential to all our treatment, and if it were not true, there would be substantial "double counting" of the effects we are discussing. The idea here is that we can add up the number of band states, expressed as spherical components at each site, to get the total number of states, as in a tight-binding approximation for the band.

From Eq. (7), we see that if $d\rho_0/dE$ is positive, the DOS increases for $E>E_m$ and decreases for $E<E_m$. This would be true for a free-electron band. In our treatment, however, we consider an inverted parabolic band, since in the vicinity of the Γ point its shape is similar to that of the p band arising from the pnictogens, with which the f electron in Ce^{3+} hybridizes[12] to give a substantial contribution to the unusually large $\langle 001 \rangle$ anisotropy. For the inverted parabolic band, the unperturbed DOS is given by

$$\rho(E)\propto(E_t-E)^{1/2}\ , \tag{8}$$

where E_t is the position of the top of the band. The DOS due to band-f mixing, as given by Eq. (7), is shown in Figs. 1–3, for different positions E_m and widths Δ_m [determined by specifying values of E_t-E_m and V for a specified unperturbed DOS, see Eq. (5)] of the local level. The unphysical behavior of the DOS (becoming negative) close to $k=0$ arises from having a constant V_k as $k\rightarrow0$, while from general considerations it can be shown[7] that the mixing vanishes at $k=0$.

When the populations $\langle n_m \rangle$ in different localized $|m\rangle$ states change as magnetic ordering occurs, E_m changes correspondingly [see Eq. (3)]. As the band states shift in

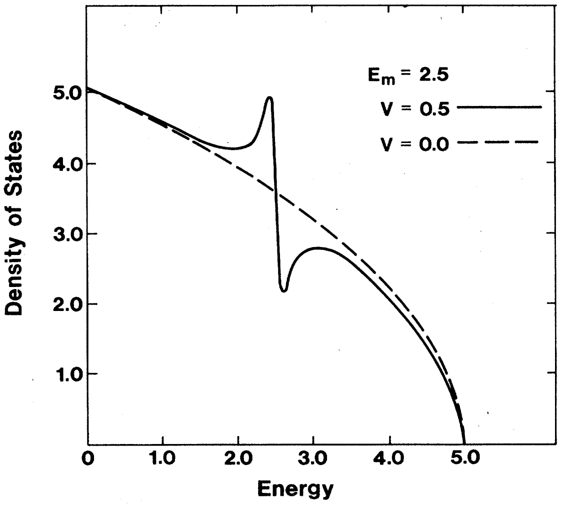

FIG. 1. Density of states for the inverted parabolic band considered in the text, with $E_t = 5.0$, $E_m = 2.5$, and $V = 0.5$. The dashed line is the density of states for the unperturbed band, shown for comparison. The energies and the density of states are in arbitrary units.

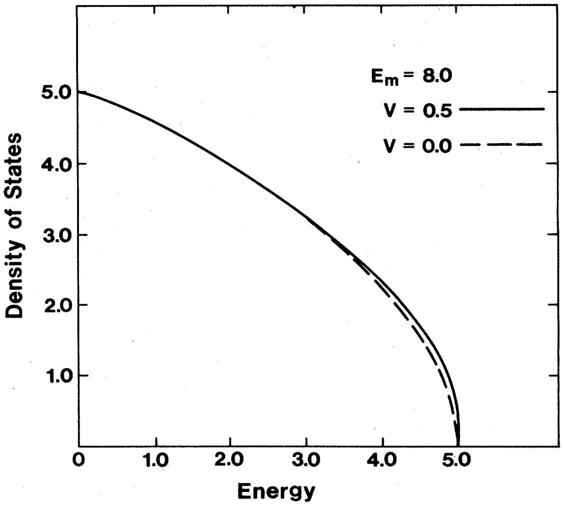

FIG. 3. Density of states for the inverted parabolic band considered in the text, for $E_m = 8.0$ and $V = 0.5$. The dashed line is the density of states for the unperturbed band, shown for comparison. The energies and the density of states are in arbitrary units.

energy around the local level, for different m this introduces unequal changes in the band DOS at a given energy. The populations of the bands are rearranged, and the resulting polarization depends on the shape of the band, the mixing strength V, the position of the local level E_m, and the unperturbed Fermi level E_F^0. From Figs. 1—3, we see that the DOS can be enhanced at certain energies and depressed at others. For a well-chosen set of parameters, the DOS can be substantially enhanced at the Fermi level (which has to be recalculated) for those m values of interest, $+\frac{1}{2}$ and $-\frac{1}{2}$ with respect to the interionic axis, on which the two-ion interaction depends.[1,2,5]

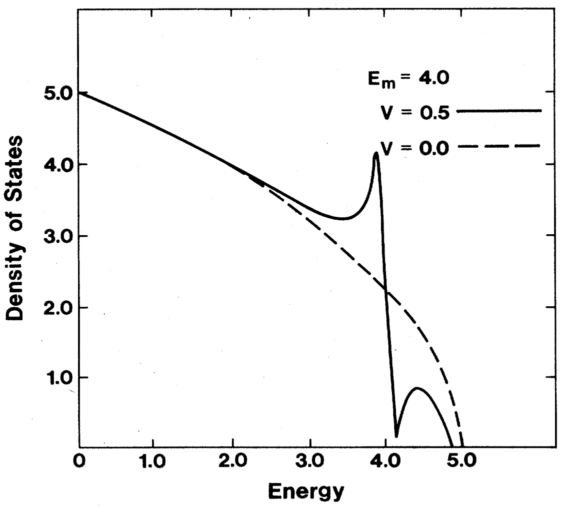

FIG. 2. Density of states for the inverted parabolic band considered in the text, for $E_m = 4.0$ and $V = 0.5$. The dashed line is the density of states for the unperturbed band, shown for comparison. The energies and the density of states are in arbitrary units.

B. Polarization and effect on two-ion interaction

The polarization which occurs through this mechanism can be substantial, since there can be large changes in the DOS as seen from Figs. 1—3. As seem from Eq. (7), the polarization depends sensitively on the position of the localized level, and on the unperturbed density of states (since the changes depend on $d\rho_0/dE$). For a constant density of states, there would be no polarization.

Starting from the Hamiltonian $\mathscr{H}$ given by the Eqs. (1) and (2), we consider $\mathscr{H}_1$ as a perturbation on $\mathscr{H}_0$ and apply the Schrieffer-Wolff transformation, to get[10,11] the single-site hybridization Hamiltonian H_h at any ionic site,

$$\mathscr{H}_h = -\sum_{\substack{kk'\\ mm'}} J_{kk'} c^{\dagger}_{k'm'} c_{km} c^{\dagger}_{m} c_{m'} \,, \tag{9}$$

with

$$J_{kk'} = \frac{V_k V^*_{k'}}{2} U \left[\frac{1}{(\epsilon_k - E_0)(\epsilon_k - E_0 - U)} + \frac{1}{(\epsilon_{k'} - E_0)(\epsilon_{k'} - E_0 - U)} \right] . \tag{10}$$

Here m,m' denote the magnetic quantum numbers of the one electron states of the localized f level (f^1) and the band.

The band-electron mediated two-ion interaction is obtained by taking the hybridization to second order,

$$\mathscr{H}_{12}(R) = \frac{\langle k\sigma | H_h^{(2)} | k'\sigma' \rangle \langle k'\sigma' | H_h^{(1)} | k\sigma \rangle}{\epsilon_{k\sigma} - \epsilon_{k'\sigma'}} \,, \tag{11}$$

where R is the vector joining sites 1 and 2. The interaction is obtained by summing over all occupied band states $|k\sigma\rangle$ and over all empty band states $|k'\sigma'\rangle$. The two-ion coupling strength for the process which changes the magnetic quantum numbers on one ion from m to m' and on the other from m' to m, is

$$E^{mm'}(R)=\sum_{\substack{k\sigma\\ k'\sigma'}}\frac{f_{k\sigma}(1-f_{k'\sigma'})}{\epsilon_{k\sigma}-\epsilon_{k'\sigma'}}\,|J^{mm'}_{k\sigma k'\sigma'}|^2 e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} \tag{12}$$

where ϵ_{ks} is the energy of the band state $|ks\rangle$, f_{ks} is the Fermi function. $J^{mm'}_{k\sigma k'\sigma'}$ involves $J_{kk'}$ given by Eq. (10), and the decomposition of the spherical waves $|km\rangle$ (about the single site) into band states $|k\sigma\rangle$; this is given by

$$J^{mm'}_{k\sigma k'\sigma'}=J_{kk'}\langle k\sigma\,|\,km\rangle\langle k'm'\,|\,k'\sigma'\rangle\;. \tag{13}$$

It is seen from the Eq. (12) that the important contribution to the summation over the $k\sigma$ and $k'\sigma'$ states comes from the states at the Fermi energy. For unpolarized free-electron bands, Eq. (12) can be simplified by integrating over k, keeping $k-k'=q$ constant, to get

$$E^{mm'}(R)=-\rho(E_F)\frac{1}{2N}\sum_{q}|J^{mm'}(q)|^2 f(q/2k_F)e^{i\mathbf{q}\cdot\mathbf{R}}\;, \tag{14}$$

where $\rho(E_F)$ is the density of states at the Fermi energy, k_F is the Fermi wave vector, and $f(x)$ is the Lindhart dielectric function[13] for zero-energy transfer,

$$f(x)=1+\frac{1-x^2}{2x}\ln\left|\frac{1+x}{1-x}\right|\;. \tag{15}$$

For $J(q)$ independent of q, the summation over q can be performed analytically to yield the RKKY interaction.[3]

In the present problem, the nature of the bands and the q dependence of $J(q)$ are not known. However, the result that the interaction strength $E^{mm'}(R)$ is proportional to the density of states at the Fermi energy is expected to remain true. Also, on performing the summation over states, it is possible to show[5] that for spherical potential mixing, the predominant interaction between two ions separated by a distance $|R|\gg k_F^{-1}$ (which is the case in these compounds) comes from the $m_l=0$ part of the scattering from each ion, so that for obtaining the two-ion interaction, only $m,m'=\pm\frac{1}{2}$ are important. We shall assume that the changes in the two-ion interaction due to the polarization are sufficiently small so they do not invalidate this important result.

As the predominant two-ion coupling occurs for $m_l=0$, we are interested in the DOS at the Fermi energy for $m_l=0$. When the bands are polarized, the interaction strengths for $m=m'=\frac{1}{2}$, $m=m'=-\frac{1}{2}$, and $m=-m'=\pm\frac{1}{2}$ scattering events are all different, as the summations over the bands in Eq. (12) now depend explicitly on m,m' due to the changes introduced by the polarization in the dispersion for up and down spins [for $m,m'=\pm\frac{1}{2}$, we have $m=\sigma$, and $m'=\sigma'$ in Eq. (12) and Eq. (13)]. In Eq. (11) we now have to introduce the first-order (in J) changes in the band eigenstates as well as the change in the energy denominator. These effects have not yet been calculated exactly. However, to make a comparison between the situations with and without the polarization effect, we adopt the following procedure. We assume that in calculating the interaction strength $E^{mm'}(R)$, the range factor which depends on $|R|$ remains unchanged (this function is a phenomenological parameter in our theory,[1,2,4,6] and is not calculated explicitly), but we shall calculate the change in the angular dependence of the interaction due to the explicit dependence of the DOS on m and m'.

When $m=m'$ the summation over band states in Eq. (12) involves only one direction of spin (either up or down), and irrespective of the dispersion relation for the band, the angular dependence is proportional to $\rho_m(E_F)$, the DOS at the Fermi level for the band of magnetic quantum number m, as only the states at E_F are important. In this case, the contribution from the terms in $f_{k\sigma}f_{k\sigma}$ add up to zero.[3] But when $m\neq m'$, the integration over the band becomes complicated, as the cross terms in $f_{k\sigma}f_{k'\sigma'}$ contribute, and their contribution has not yet been calculated. In the absence of an exact calculation, we assume the result to be proportional to $[\rho_m(E_F)\rho_{m'}(E_F)]^{1/2}$. This function gives the correct results in the unpolarized case, and goes to zero in the limits of the DOS for either band going to zero (i.e., if there is zero density of states at the Fermi level for either band, the interaction between ions does not exist). The ratio of the interaction strength in the presence, to that in the absence, of polarization is then

$$\frac{E^{mm'}}{E_0^{mm'}}=\frac{[\rho_m(E_F)\rho_{m'}(E_F)]^{1/2}}{\rho_0(E_F^0)} \tag{16}$$

where 0 refers to the unperturbed band states.

In the following discussion we assume a ferromagnetically ordered system; we shall choose the quantization axis to lie along the moment direction. We assume that the density of band states for all m values are equal in the absence of the hybridization interaction. For a saturated moment, $\langle n_{5/2}\rangle=1$ and $\langle n_m\rangle=0$ for $m\neq\frac{5}{2}$, for quantization along the moment direction. The projection of a state $|m\rangle$ on to a general direction of the interionic axis as quantization axis is achieved by using the rotational transformation to that axis.

The band DOS for $m,m'=\pm\frac{1}{2}$ along the interionic axis in the presence of band distortion for the saturated ferromagnetic moment case has been calculated numerically for the inverted parabolic band whose unperturbed DOS is given in Eq. (8). This also involves a recalculation of the Fermi energy, which has changed due to the rearrangement of the populations. Using Eq. (16), the ratio of the new and old interaction constants for a given direction can be found. The change in the angular dependence of the interaction from the case in which band distortion is not taken into account, itself depends on the angle between the moment direction and the interionic axis. Thus the band distortion effect changes the dependence of the free energy on the direction of moment.

We have investigated the effect of the distortion of the

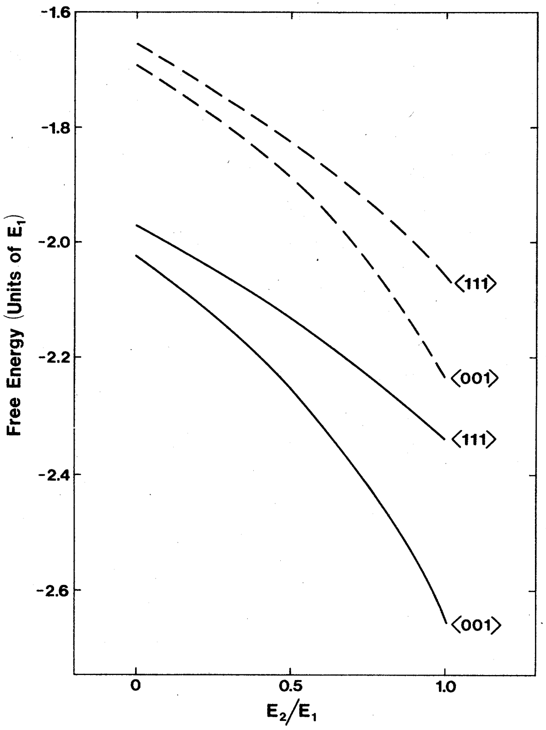

FIG. 4. Free energies of the ferromagnetic states with moments along the ⟨001⟩ and ⟨111⟩ directions, for varying ratios of NN and NNN ferromagnetic hybridization-mediated two-ion interaction strengths E_2/E_1, for the situations in which the band distortion effect has been included (solid lines) and has not been included (dashed lines). Parameters are as given in the text.

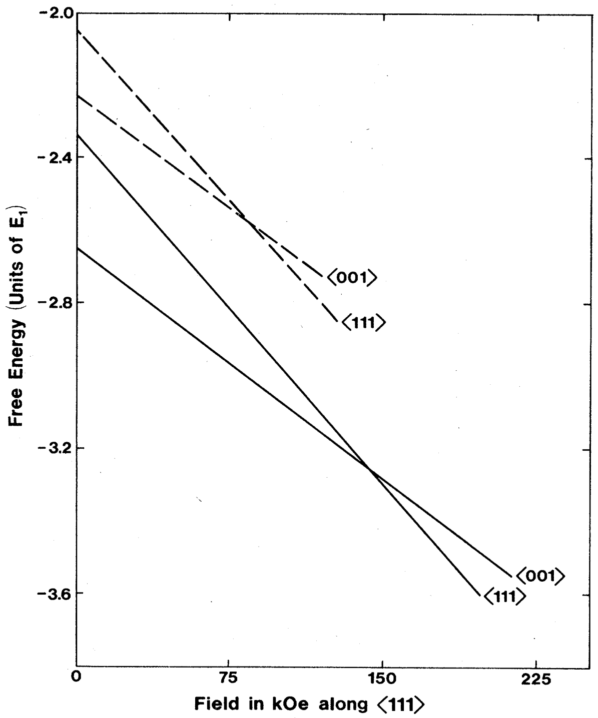

FIG. 5. Free energies of the ferromagnetic states with moments along the ⟨001⟩ and ⟨111⟩ directions, for $E_2=E_1=|E_1|$, as a function of an external magnetic field applied along ⟨111⟩, for the situations in which the band distortion effect has been included (solid lines) and has not been included (dashed lines). Parameters are for a case modeling CeBi as given in the text.

DOS of the band on the ferromagnetically ordered f^1 case, for the cube-edge and the body-diagonal directions of saturated moment. Figure 4 shows the free energies of these ferromagnetic states, that are obtained for ferromagnetic nearest-neighbor (NN) and next-nearest-neighbor (NNN) hybridization-mediated Coqblin-Schrieffer (CS) interactions, for varying E_2/E_1 (E_n is the CS interaction strength to the nth near neighbor), for the cases where the associated changes in the band structure have not and have been included. At $E_2=E_1=|E_1|$, the ordered ferromagnetic moment is over 99% saturated, but as (E_2/E_1) is reduced, the moment moves away from saturation[14] almost linearly to about 72% of the saturation moment at $E_2=0$. Here, we have assumed saturated ionic moments (hence the accuracy of the calculations shown in Fig. 4 decreases with decreasing E_2/E_1), and the parameters $E_t=5$ eV, $E_F=4.5$ eV, $E_0=1$ eV, $U=5$ eV, $V=0.5$ eV were chosen [the various E_m and Δ_m can be calculated from these parameters using Eqs. (3) and (8) in Eq. (5)]. (These parameters were chosen so as to increase the DOS of $m=\pm\frac{1}{2}$ components of the bands at the Fermi energy. Not all parameter sets increase these DOS from the unperturbed values; some of them may decrease the DOS.) The band-distortion effect increases the magnitude of the free energies of both the ⟨001⟩ and ⟨111⟩ ferromagnets (makes both more negative); but as the ratio E_2/E_1 is increased, this effect relatively favors the ⟨001⟩ direction of moment.

For the ferromagnet of Fig. 4, when the band distortion is not considered, for equal NN and NNN CS interaction an ⟨001⟩ alignment of moment is preferred; and a magnetic field of about 80 kOe applied along ⟨111⟩ is required to change the direction of moment from ⟨001⟩ to ⟨111⟩. Figure 5 shows the free energy changes giving this change in direction of moment alignment for this $E_1=E_2$ case. Adding a small antiferromagnetic isotropic interaction stabilizes the type-I and type-IA antiferromagnetic structures in zero external field, with ordering temperatures as in[1,2,5] CeBi. Thus the model's behavior is characteristic of CeBi. Corresponding to the favoring of ⟨001⟩ alignment shown in Fig. 4, the polarization (band-distortion) effect increases the field required to switch the moment alignment from ⟨001⟩ to ⟨111⟩; and in Fig. 5 we see that in our case modeling CeBi, this critical field is increased from about 80 to over 140 kOe. [We assume that the polarization effects are important only well below the ordering temperature T_0, and that the polarization effect does not significantly alter the transition temperature at which the material becomes magnetically ordered (this would be more likely to be true for a second-order transition, rather than a first-order transition).] This value for the anisotropy field is still smaller than what is experimentally observed. To increase the anisotropy field further, one would have to resort to larger values of the mixing strength V. However, in that case, the perturbation theory giving the two-ion interaction, as has been used here, may not be valid.

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