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Calculation of magnetic impurities in a nonmagnetic host: Fe in Au

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The fully relativistic spin-polarized embedded-cluster method is discussed for magnetic impurities in nonmagnetic host metals. We apply the method to the calculation of the density of states and the magnetic energy of the cases of a single Fe impurity in Au, and of a pair of Fe atoms located within a first-neighbor shell cluster in Au. In addition to allowing for different distances between the two Fe impurities, we also allow for different spin orientations, with respect to the lattice symmetry of the host. We have also analyzed the spin polarization induced on a central Au site by the Fe atom(s). Thus, we have shown that the method is appropriate to obtain effective spin Hamiltonians for local moment systems, which contain local anisotropy terms and anisotropic exchange interactions.

I. INTRODUCTION

Materials composed of either transition metals or actinide elements are sources of a very rich variety of interesting and unusual phenomena. Electronic structure calculations have proven to be extremely useful in the understanding of their electronic properties. However, one of the outstanding problems remaining is that of describing materials in which the nonlinearity due to electron-electron interactions becomes comparable to the dispersive character of the one-electron excitations. In many such cases, the solution of the full, nonlinear, Schrödinger equation is no longer unique due to the existence of states with spontaneously broken symmetry.[1] A common form of spontaneous symmetry breaking found in transition metals and components is magnetic ordering,[2] and similarly moment formation of impurities in nonmagnetic hosts.[3] Electronic structure calculations including spin polarization[4-7] are essential in the description of such materials.[8-10] Since the existence of spin is a relativistic phenomenon, a derivation of the spin-polarized Kohn-Sham equations must be based on a fully relativistic density-functional theory. Rajagopal and Callaway,[11] obtained a spin-polarized theory as the nonrelativistic limit of a fully relativistic theory. However, the appearance of a fully relativistic, spin polarized, theory[12,13] is a relatively new development that was prompted by the growing interest in the actinide materials,[14,15] where spin-orbit and spin-polarization effects are large. These developments allow for a rigorous treatment of spin only magnetism in f-electron systems.[16] This development also enables rigorous investigations of subtle effects such as magnetic anisotropy,[17] in which the spin polarization couples to the lattice via the spin-orbit interaction, and requires both effects to be treated on the same footing.

The existence and stability of local magnetic moments in nonmagnetic hosts[3,18-20] is central in our understanding of many physical phenomena, such as giant local moments,[21-23] spin glasses,[24-26] and even magnetic ordering in some concentrated alloys and compounds.[27-29] The stability of local moments is often attributed to intra-atomic correlations. As an example, a commonly used model for moment instability is the Kondo effect,[3,30,31] which is appropriate when intra-atomic correlations restrict the wave functions to be a linear combination of wave functions corresponding to a degenerate and a nondegenerate configuration. On the other hand, a linear combination corresponding to two distinct degenerate configurations is assumed to lead to the formation of a stable local moment.[32] This simple model does fail to attach much significance to the nature of the host's conduction band and therefore accounts poorly for the appearance of local moments in some hosts, but not others. In fact, the qualitative differences between hosts with *sp* conduction bands and *d* bands remain unexplained.[19] The importance of the host polarizability is evidenced by $3d$ impurities in Pd,[21-23] in which the host bands exhibit a large local spin polarization giving rise to giant moment formation. The polarizations induced in the conduction bands by the interaction with the polarized impurity, also forms an essential part to the description of a coupling between local moments[17,27] as for example modeled by the Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interactions. A proper description of the single-site spin anisotropy,[33,34] and the charge anisotropy[23,29] requires a simultaneous inclusion of both spin-polarization and spin-orbit couplings at the sites of the local moments, and in the host conduction bands, as well as their mutual interactions.

In this paper, we address the problem of a stable spin-polarized $3d$ impurity in a nonmagnetic host and the interactions between two such impurity whether direct or mediated by the polarization of the intervening nonmagnetic host atoms. We treat this problem within the framework of density functional theory. Since these calculations are not only based on a new type of approach but also are computationally quite intensive, we do not perform one-electron charge-density self-consistent calculations. Therefore, one ought to consider our results as model calculations from which, we hope, some insight may be gained.

In Sec. II, we discuss the fully relativistic, spin-polarized, embedded-cluster method. In order to reduce the size of this section, we have only briefly outlined the theoretical results.[12,13,16,35] In Sec. III, we present a short account of the computational details. In Sec. IV, we show the results for one impurity and two impurity Fe atoms in an Au host metal. We also discuss the effects of the polarization of the Fe atoms on an Au site. Our results for the single impurity case shows that the qualitative, interpretative picture of "spin-up" and "spin-down" densities of states (DOS) is not automatically transferrable to the concept of spin only (m_s-) projected DOS in a relativistically spin-polarized approach.

II. METHOD OF CALCULATION

In order to describe the scattering from more than one impurity, one needs to calculate the so-called site off-diagonal Green's functions $G(\mathbf{r},\mathbf{r}')$, i.e., Green's functions, where the two spanning $\mathbf{r}$ and $\mathbf{r}'$ are measured from two different lattice sites. In that case, the off-diagonal scattering path operator for a nonmagnetic cubic lattice with one atom per unit cell is given by,[35]

$$\begin{aligned}\underline{\tau}^{ij}(\varepsilon)&=2\Omega_{\mathrm{IBZ}}^{-1}\sum_{S\in O}\int_{\Omega_{\mathrm{BZ}}}\underline{D}^{\dagger}(S)\underline{\tau}(\mathbf{k},\varepsilon)\underline{D}(S)\cos[\mathbf{k}\cdot S(\mathbf{R}_i-\mathbf{R}_j)]d^3k\\&=\Omega_{\mathrm{IBZ}}^{-1}\int_{\Omega_{\mathrm{BZ}}}[t_0^{-1}(\varepsilon)-G(\mathbf{k},\varepsilon)]^{-1}e^{i\mathbf{k}\cdot(\mathbf{R}_i-\mathbf{R}_j)}d^3k\\&\mathbf{R}_i,\mathbf{R}_j\in L;\mathbf{k}\in L_0^{-1};O\subset O_h\ ,\end{aligned}\tag{1}$$

where Ω_{BZ} is the volume of the Brillouin zone (BZ), Ω_{IBZ} the volume of the irreducible wedge of the BZ, $t_0(\epsilon)$ is the single site t matrix of the host, and $G(\mathbf{k},\varepsilon)$ are the corresponding structure constants. In (1) underbars denote matrices (e.g., angular momentum representation) and the matrices $\underline{D}(S)$ contain blockwise the Clebsch-Gordan coefficients for the irreducible projective representations[36] of the cubic group O_h. Using these representations only 24 BZ integrals need to be calculated for O instead for the usual 48 of the double group.

For the problem of several impurities (embedded cluster) one constructs a supermatrix $\underline{\underline{\tau}}(\varepsilon)$ with elements $\underline{\tau}^{ij}(\epsilon)$.

$$\underline{\underline{\tau}}(\varepsilon)=\begin{bmatrix}\underline{\tau}^{ii}(\varepsilon) & \underline{\tau}^{ij}(\varepsilon)\\ \underline{\tau}^{ji}(\varepsilon) & \underline{\tau}^{jj}(\varepsilon)\end{bmatrix}\quad i,j=1,N\ ,\tag{2}$$

where N is the number of embedded sites. For each site i in the embedded cluster a local transformation, e.g., $\underline{D}_i(R)$ can be considered. For the whole cluster these transformation are conveniently arranged in supermatrices $\underline{\underline{R}}$ and $\underline{\underline{R}}^{-1}$ (inverse), i.e.,

$$\begin{gathered}\underline{\underline{R}}=\begin{bmatrix}\underline{D}_i(R) & 0 & 0\\ 0 & \underline{D}_j(S) & 0\\ O & 0 & \underline{D}_k(T)\end{bmatrix},\\ \underline{\underline{R}}^{-1}=\begin{bmatrix}\underline{D}_i(R^{-1}) & 0 & 0\\ 0 & \underline{D}_j(S^{-1}) & 0\\ O & 0 & \underline{D}_k(T^{-1})\end{bmatrix}.\end{gathered}\tag{3}$$

Turning now to the problem of magnetic impurities, one constructs a diagonal supermatrix, whose elements are given by the inverse single-site t matrices. If sites i and j are occupied by impurities, the elements i and j of this supermatrix are given by the inverse magnetic single-site t matrix[12,13,16] of the impurity species α, whereas all other elements are given by the inverse single-site t matrix of the host which, since they are diagonal angular representations, are invariant under transformations:

$$\underline{t}^{-1}(\underline{\underline{R}}^{-1},\varepsilon)=(\underline{\underline{R}}^{-1})^{\dagger}\underline{t}^{-1}(\varepsilon)\underline{\underline{R}}^{-1}$$

$$=\begin{bmatrix} \underline{t}_0^{-1}(\varepsilon) & 0 & 0 & 0 \\ 0 & \underline{D}_i(R)\underline{t}_{i\alpha}^{-1}(\varepsilon)\underline{D}_i(R) & 0 & 0 \\ 0 & 0 & \underline{D}_j(S)\underline{t}_{j\alpha}^{-1}(\varepsilon)\underline{D}_j(S) & 0 \\ 0 & O & 0 & \underline{t}_0^{-1}(\varepsilon) \end{bmatrix} . \tag{4}$$

In (4) for sites i and j the axis of rotation is rotated back to the case for which the spin-polarized Kohn-Sham-Dirac equations can be solved[12,13] (internal effective field $B_{\text{eff}}(r)$ along the Z axis). Rotation of the cluster τ matrix[35] [Eq. (2)] with $\underline{\underline{R}}$ therefore takes account of the case of different orientations on different sites:

$$\underline{\underline{\tau}}(\underline{\underline{R}},\varepsilon)=\underline{\underline{R}}^{\dagger}(\{1+[\underline{t}^{-1}(\underline{\underline{R}}^{-1},\varepsilon)-\underline{\underline{t}}_0^{-1}(\varepsilon)]\underline{\underline{\tau}}(\varepsilon)\}^{-1}\underline{\underline{\tau}}(\varepsilon))\underline{\underline{R}} . \tag{5}$$

If in (5) all sites in the embedded cluster are occupied by host atoms, $\underline{\underline{\tau}}(\underline{\underline{R}},\varepsilon)$ is given simply by a rotated cluster matrix $\underline{\underline{\tau}}(\varepsilon)$ [Eq. (2)], which of course, e.g., yields nothing but N times the density of states for the host metal. Inspecting in (5) the ith site, the scattering path operator is given by

$$\underline{\tau}^{ii}(\underline{\underline{R}},\varepsilon)=\underline{D}_i^{\dagger}(R)(\{1+[\underline{t}^{-1}(\underline{\underline{R}}^{-1},\varepsilon)-\underline{\underline{t}}_0^{-1}(\varepsilon)]\underline{\underline{\tau}}(\varepsilon)\}^{-1}\underline{\underline{\tau}}(\varepsilon))_{ii}\underline{D}_i(R) . \tag{6}$$

i.e., by the ith diagonal block in Eq. (5). This cluster-diagonal block clearly depends not only on the occupation of site i by species α and the local orientation of the internal effective field $B_{\text{eff}}(r)$, but also on the environment of the corresponding quantities. The site DOS $n^i(\underline{\underline{R}},\varepsilon)$ and the site magnetization DOS $m^i(\underline{\underline{R}},\varepsilon)$ for an impurity atom of species α occupying site i are then given by[16]

$$n^{i\alpha}(\underline{\underline{R}},\varepsilon)=-\pi^{-1}\text{Im}\,\text{Tr}[\underline{R}^{i\alpha}(\varepsilon)\tau^{ii}(\underline{\underline{R}},\varepsilon)] , \tag{7}$$

and

$$m^{i\alpha}(\underline{R},\varepsilon)=-\pi^{-1}\text{Im}\,\text{Tr}[\beta\sigma_z\underline{R}^{i\alpha}(\varepsilon)\tau(\underline{R},\varepsilon)] , \tag{8}$$

where the matrix of radial integrals $\underline{R}^{i\alpha}(\varepsilon)$ is defined in Eq. (7)–(10) of Ref. 16. As can easily be shown from (5)–(8), if only a single magnetic impurity is considered, the magnetization density and therefore the magnetic moment have the same values for all $R \in O_h$.

III. COMPUTATIONAL DETAILS

For the host metal (Au) a muffin-tin fully relativistic, self-consistent linear-muffin-tin orbital (LMTO) potential corresponding to a lattice constant of $a_0=7.681\,75$ a.u. was used (muffin-tin radius $R_s=2.585\,71$ a.u.). The BZ integrals (1) for the off-diagonal scattering path operators of the host are calculated in the complex plane parallel to the real axis using the 21 special directions of Fehlner and Vosko[37,38] and are then analytically continued to the real axis. All spectral quantities are obtained with an energy mesh of 0.01 d.u. [1 d.u.$=(2\pi/a_0)^2$ Ry].

For the Fe impurities the results of the (nonrelativistic) spin-polarized calculations of Moruzzi *et al.*[7] are shifted by a constant such that the Fermi energy of Fe matches the Fermi energy of the host metal (Au). The spin-polarized charge densities of Moruzzi *et al.*[7] are used to generate the effective potential and the effective internal field[12,13] in terms of the local spin-density functional given by Gunnarson.[39] Throughout this work a muffin-tin radius for Fe of 2.225 54 a.u. was used (which contained 7.9d electrons).

In the present paper, only the d-like quantities are shown, which are defined as the corresponding d-like contributions in Eqs. (7) and (8). Denoting the d-like magnetization DOS for a particular arrangement $\underline{\underline{R}}$ simply by $m_d(\varepsilon)$, the d-like magnetic moment by $m_d(\mu_B)$, and the corresponding calculated d-like magnetic energy by E_m^d, one obtains the relations

$$m_d=-\int_0^{E_F} m_d(\varepsilon)d\varepsilon,\quad E_m^d=-\int_0^{E_F} m_d(\varepsilon)\varepsilon\, d\varepsilon , \tag{9}$$

where E_F is the Fermi energy of the host.

IV. RESULTS AND DISCUSSION

A. Single impurities

In order to get some reference for the many impurity problems we first consider the single impurity problem.

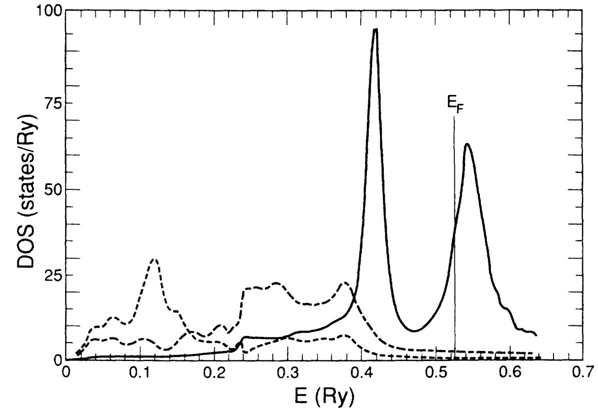

FIG. 1. Nonmagnetic $d^{3/2}$-like (dashed line) and $d^{5/2}$-like (dashed-dotted line) partial local DOS's for the host (Au) and the spin-polarized d-like DOS of a single impurity of Fe in Au (solid line).

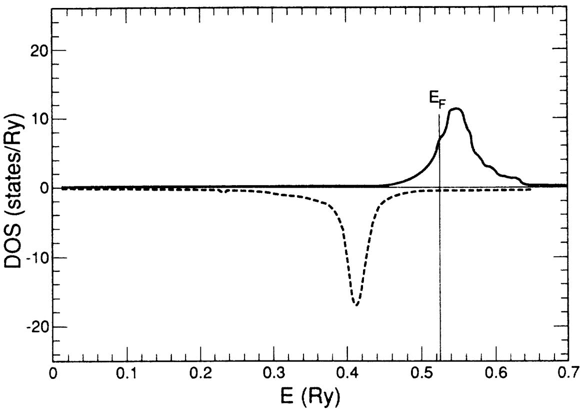

FIG. 2. $m_s=\frac{1}{2}$ (dashed line) and $m_s=-\frac{1}{2}$ (solid line) projected *d*-like DOS's for a single impurity of Fe in Au. For $m_s=-\frac{1}{2}$, (-1) times the corresponding DOS is shown.

In Fig. 1 we show the ("paramagnetic" $d^{3/2}$- and $d^{5/2}$-like DOS for the host (Au) together with the fully relativistic *d*-like ("spin-polarized") DOS for a single impurity of Fe in Au. The Fermi energy E_F of the host is at 0.528 Ry (vertical line). These results can be compared to the non-spin-polarized relativistic calculation of a single impurity of Fe in Au that shows only one large *d*-like "virtual bound state"[40] just below E_F. The energetic positions of the two peaks in the *d*-like DOS for Fe (Fig. 1) correspond to the peaks in the nonrelativistic "spin-up" and "spin-down" *d*-like densities of a single impurity of Fe in Au. For a better understanding of the origin of those two peaks, we show in Fig. 2 the $m_s=\pm\frac{1}{2}$ projection of the *d*-like DOS and in Fig. 3 the $j=\frac{5}{2}$, $\mu=\pm\frac{5}{2}$-like partial local DOS. As can be seen from Fig. 2, both the $m_s=\frac{1}{2}$ and $m_s=-\frac{1}{2}$ projections have peaks at the energetic positions of the spin-polarized virtual bound states. Since for $l=2$, the ($j=\frac{5}{2}$, $\mu=\pm\frac{5}{2}$) channels are the only ones that cannot mix,[14,17] the corresponding DOS have only one peak each, thus mimicking what one is used to seeing in terms of a nonrelativistic spin-polarized calculation for the *d* channels (at least in the case of rather large "exchange splitting"). The *d*-like DOS is given by the sum of the $m_S=\frac{1}{2}$ and $m_S=-\frac{1}{2}$ projected local DOS, whereas the *d*-like magnetization DOS is given by their difference. It should be noted that all the other *d*-like partial local DOS for a single impurity of Fe in Au, namely $j=\frac{3}{2},\frac{5}{2}$; $\mu=\pm\frac{1}{2},\pm\frac{3}{2}$, do have two peaks each located energetically in the vicinity of the two spin-polarized *d*-like virtual bound states.

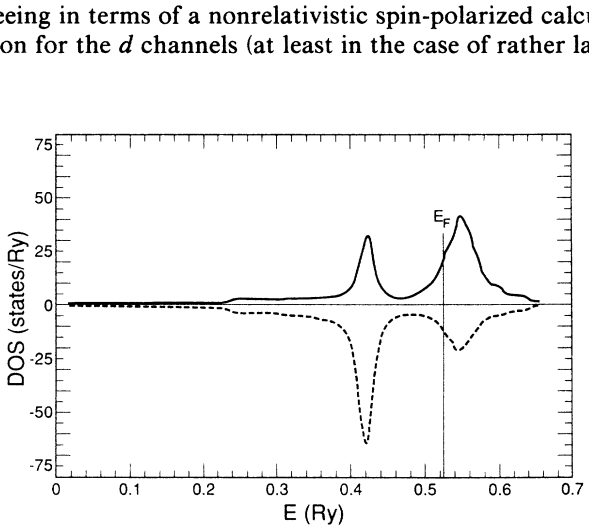

FIG. 3. ($j=\frac{5}{2}$, $\mu=\pm\frac{5}{2}$) *d*-like DOS's for a single impurity of Fe in Au. For $\mu=\frac{5}{2}$ (dashed line) (-1) times the corresponding partial local DOS is shown.

The *d*-like magnetic moment obtained by integrating the *d*-like magnetization DOS up to the Fermi energy of the host is $3.1259\mu_B$ and the corresponding magnetic energy is 1.1312 Ry. As already mentioned in the preceding section the magnetic moment for a single magnetic impurity in a (paramagnetic) cubic host is independent of applied rotations for the "orientations of the spin."

An interesting question to ask is, how does the magnetic moment go to zero if the internal effective field is scaled to zero. This addresses the question of how the nonmagnetic limit is approached within a spin-polarized relativistic theory. In Fig. 4 the *d*-like magnetic moment and the *d*-like magnetic energy of a single impurity of Fe in Au is shown as a function of a scaling factor x (extending from zero to 1) for the internal effective field. As one can see from the curve for the *d*-like magnetic energy one indeed gets an asymptotic behavior for $x \rightarrow 0$. However, one can see also that for values of x near 1.0 the shape of the curve is by no means linear in x. Beyond the question

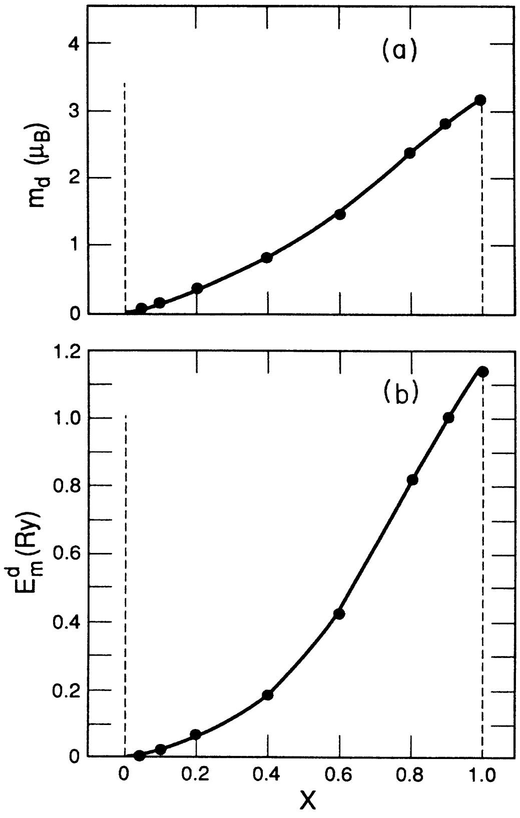

FIG. 4. (a) *d*-like magnetic moment (μ_B) and (b) *d*-like magnetic energy (Ry) for a single impurity of Fe in Au as a function of the applied internal field $B_{\text{eff}}(r)$ with respect to a scaling factor x.

of the nonmagnetic limit, Fig. 4 is also of practical interest, since during a self-consistent spin-polarized calculation the d-like magnetic moment moves up and down in the vicinity of $x=1$ until it has reached its final, i.e., self-consistent, value.

B. Double impurities

Having more than one impurity opens up new possibilities. By placing two impurities of Fe in Au, the magnetic moment and therefore also the magnetic energy is no longer invariant with respect to local rotations of the internal field. Within the fcc unit cell there are four inequivalent (nonzero) distance vectors $\mathbf{R}_{ij}=\mathbf{R}_i-\mathbf{R}_j$ between the two impurity sites. The two magnetic Fe atoms can therefore be separated by four different distances [nearest neighbor (NN), second nearest neighbor (SNN), etc.]. Since in the formulation of the spin-polarized relativistic Kohn-Sham-Dirac equations[17,37] the effective field $\mathbf{B}_{\rm eff}(r)$ is assumed to point along the Z axis, the magnetization DOS $m(E)$ is invariant under rotations $R\ (\in O_h)$ around the Z axis. Therefore choosing an axis of rotation perpendicular to the Z axis the local Fe rotations leads to an orientation dependent magnetization DOS per site. Throughout this paper rotations around the X axis are used, where the symbols E, C_{1X}, C_{2X}, and C_{3X} denote clockwise rotations around this axis by $0\pi(2\pi)$, $\pi/2$, π, and $3\pi/2$, respectively. It should be noted that by choosing the Y axis as the axis of rotation exactly the same results are obtained for C_{1Y}, C_{2Y}, and C_{3Y}. In the following the notation (R_1,R_2), R_1, $R_2\in O_h$ is adopted to specify the local rotations for impurity site 1 and 2.

In Fig. 5 the d-like magnetic energy per site is shown for the NN distance of two Fe atoms in a host lattice of unpolarized Au atoms. The level of reference (zero of energy) is with respect to the d-like magnetic energy of a single impurity of Fe in Au. For the cases (E,E) and (C_{2X},C_{2X}) the internal field vectors are pointing into or out of the XY plane, respectively. For the other two ferromagnetic configurations $[(C_{1X},C_{1X})$ and $(C_{3X},C_{3X})]$ the internal field vectors are pointing in the $+Y(-Y)$ directions in the XY plane. For the antiferromagnetic configurations (E,C_{2X}) and (C_{1X},C_{3X}) the internal field vectors are pointing in the $+Z$ and $-Z$ directions or in the $+Y$ and $-Y$ directions. For the two intermediate cases we considered, namely (E,C_{1X}) and (E,C_{3X}), the internal field vectors are pointing in the $+Z$ for the central atom and in the $+Y(-Y)$ directions for the corner atom. First, it is seen that the two antiferromagnetic configurations are highest in energy and differ little from the magnetic energy per site of the single impurity case. Next, the two intermediate cases have lower energy, but are not equal as the case (E,C_{1X}) has a lower energy than the case (E,C_{3X}). Finally the ferromagnetic configurations have the lowest energy of all.

In Fig. 6 we vary not only the orientations of the spin of the two Fe atoms with respect to each other but also the distance between them. Again Fig. 6 depicts the d-like magnetic energy per site of a double impurity of Fe in a host lattice of unpolarized Au atoms, with the zero of energy being the d-like magnetic energy of a single impurity of Fe in Au. As seen from Fig. 6 the smallest deviation from the d-like magnetic energy of a single impurity is for the antiferromagnetic case (E,C_{2X}). Concentrating in Fig. 6 on this case, one finds that this particular deviation goes to zero when the distance is increased beyond $1a_0$. This indicates that with increasing distance the "antiferromagnetic-like" interaction becomes weaker and each impurity behaves like a single impurity. For the other cases it is quite different. Taking for example the case of "ferromagnetic" behavior (solid circles), we see that the magnetic energy oscillates with respect to the single impurity case. One interesting aspect seen in Fig. 6 is that the lowest magnetic energy state as a function of

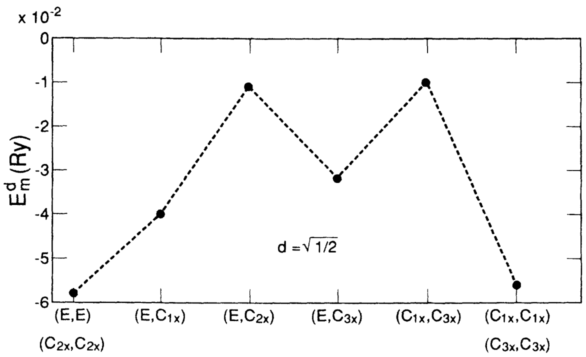

FIG. 5. d-like magnetic energy per site (Ry) for a double impurity of Fe in Au at nearest-neighbor distance with respect to rotations around the x axis. The zero of energy refers to the d-like magnetic energy of a single impurity of Fe in Au. The symbols in parentheses denote the local rotations for impurities 1 and 2.

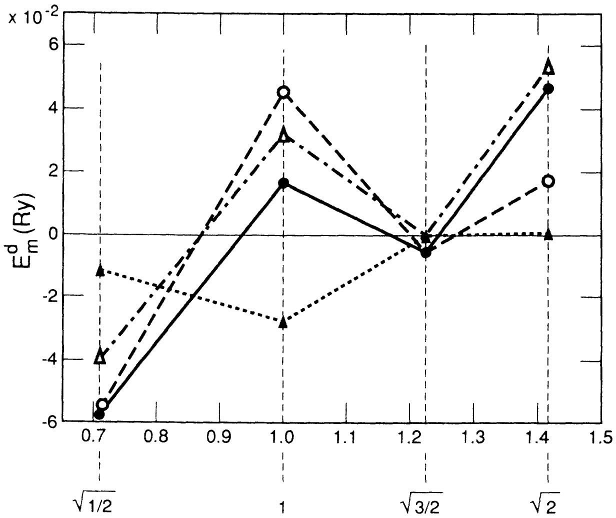

FIG. 6. d-like magnetic energy (Ry) of a double impurity of Fe in Au with respect to the distance d separating of the two Fe sites. The zero of energy refers to the d-like magnetic energy of a single impurity of Fe in Au. Solid circles: (E,E), (C_{2X},C_{2X}); open circles: (C_{1X},C_{1X}); solid triangles: (E,C_{2X}); open triangles: (E,C_{1X}).

the distance is "ferro," "antiferro," "ferro," "antiferro," suggestive of the oscillations in the RKKY exchange interaction.

C. Polarization effects on Au sites

In order to discuss polarization effects on Au sites the following procedure is used: One Au atom is spin-polarized by a very small constant effective field (-1.0×10^{-4} Ry). This means that we are breaking the crystal symmetry at this point and allow for directional exchange. Considering this spin-polarized Au atom as single impurity in a host of unpolarized Au atoms such an impurity carries a d-like magnetic moment of $0.000\,714\mu_B$ with a corresponding d-like magnetic energy of 0.000 262 Ry. This single spin-polarized Au atom serves as zero of energy for a discussion of polarization effects on an Au site induced by Fe atoms.

In order to study polarization effects by a single Fe impurity onto an Au site the case of a NN spin-polarized Au atom with all other Au atoms being unpolarized is considered. The results of these calculations are shown in Fig. 7 with respect to the local spin orientation for the Fe atom and an orientation of E for the Au atom. In Fig. 7 therefore the symbols E, C_{1X}, and C_{2X}, refer to the local spin orientations for the Fe atom. As one can see the effects of polarization on the Au site by the Fe atom decrease as the Fe atom is moved away from the Au atom under consideration. We also find that regardless of the distance, the C_{2X} orientation of the spin at the Fe site induces the lowest magnetic energy at the Au site with respect to the single spin-polarized Au impurity. If the orientations of the spin at each impurity is the same, namely E at the Au site and E at the Fe site, we find that only at a distance of 1 (solid circles) do we get a negative deviation from the single Au impurity case. Since all these calculations are non-self-consistent, we have to be careful not overstating the effects; in all considered cases the deviations from the single Au impurity case are rather small. Concentrating on the Fe site, where as mentioned previously the invariance under rotations $R\in O_h$ is now lifted because there is one polarized Au atom destroying this symmetry, we find for the orientations E, C_{1X}, and C_{2X} at the Fe site a very small decrease of the moment of Fe by 0.000 014, 0.000 049, and $0.000\,035\mu_B$, respectively.

Finally, for a discussion of polarization effects on an Au site by two impurities of Fe the case with a spin-polarized Au NN atom and two SNN Fe atoms is considered. The orientations of the Fe atoms are shown as entries in Fig. 8. In both cases the ferromagnetic interaction between the Fe atoms and the Au atom (E,E) shows an increase in the magnetic energy, whereas the antiferromagnetic configuration (C_{2X},C_{2X}) shows a decrease. Similar to Fig. 7 we find that the (C_{2X},C_{2X}) orientation for the Fe impurities seems to be favorable for both distances.

From Figs. 7 and 8 it is evident that the polarization effects on an Au site indeed not only depend on the magnitude of the inducing magnetic moment, but also on its orientation and on the distance between the inducing site and the Au site. As compared to the magnetic moment per site for Fe the induced moment on the Au site is very

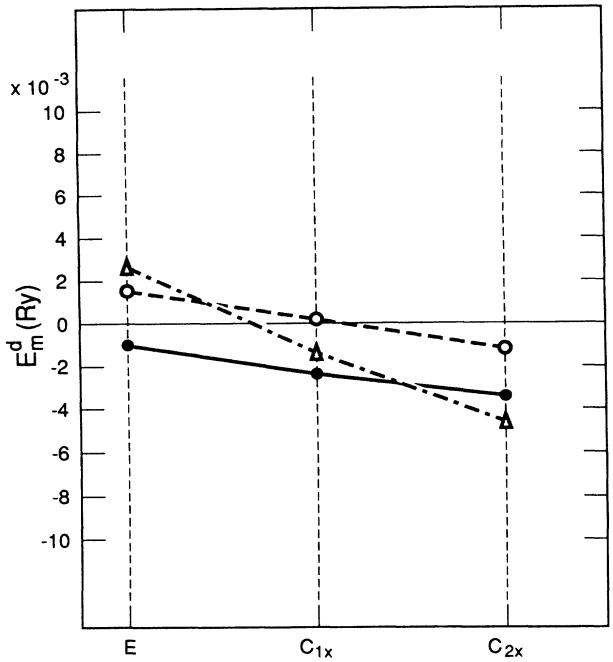

FIG. 7. Polarization effects on an Au site induced by a single impurity of Fe in Au: d-like magnetic energy (Ry) of Au with respect to the local rotation for the Fe site. Distances (a_0): triangles $\sqrt{\frac{1}{2}}$; solid circles 1; open circles $\sqrt{2}$.

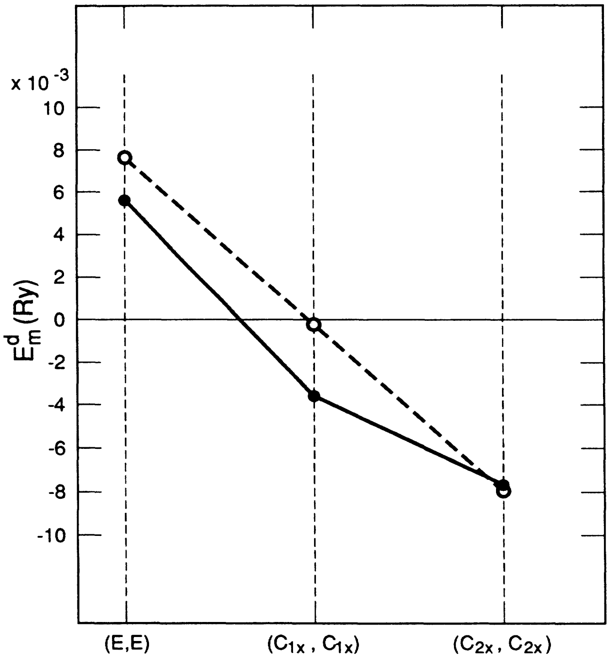

FIG. 8. Polarization effects on an Au site induced by a double impurity of Fe in Au: d-like magnetic energy (Ry) of Au with respect to the local rotations of the Fe sites. Distances (a_0): solid circles 1; open circles $\sqrt{2}$.

small, but not negligible. In fact an "academic case" of 12 Fe atoms with orientations E, i.e., all "spins lined up," occupying the first shell of neighbors in an Au host induce a magnetic moment of $-0.103\mu_B$ for a spin-polarized Au atom at the origin of this embedded cluster.

V. CONCLUSION

The present calculations serve as an illustration of the complexity to be encountered when dealing with magnetic interactions in a real system on a microscopic level. These calculations are for many reasons incomplete. First, each case considered should be performed self-consistently. Second, to study the decay of the magnetic interactions with respect to increasing distances between the magnetic sites much larger embedded clusters have to be considered. In view of these arguments the present calculations are nothing but model calculations. However, considering the various computational steps involved such as calculating the off-diagonal scattering path operators by Brillouin zone integrations and then the computation of all the various cases shown already implies considerable computational effort.

The method is neither restricted to nonmagnetic host metals nor is it restricted to pure systems. Using again group theory rather extensively, the τ^{ij} of a magnetic host (alloy) can be calculated fully relativistically and spin polarized. Of course also results of fully relativistic Korringa-Kohn-Rostoker calculations within the coherent potential approximation (KKR-CPA) can be used as input to deal with the problem of magnetic impurities in (nonmagnetic) substitutionally disordered alloys. In a different context, such fully relativistic embedded cluster calculations for substitutionally disordered alloys[41] have proven to give a highly accurate account for environment dependent physical properties.

Within the limits of these calculations we draw the following conclusions regarding the behavior of Fe impurities in Au with realistic interactions. First, the magnetic moment of the single Fe impurity in Au is larger than that of pure bulk Fe. Second, the preferred coupling between two Fe impurities oscillates as a function of the distance between the impurities (restricted to fcc lattice sites). For the nearest-neighbor distance the coupling is ferromagnetic and goes antiferromagnetic at the next-nearest-neighbor distance. Also the antiferromagnetic interaction goes to zero beyond the next-nearest-neighbor distance, whereas the ferromagnetic interaction persists. However, the coupling between a host Au atom and one or two Fe impurities is always antiferromagnetic.

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