Exchange Coupling and Conduction-Electron Polarization in Metals. II

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(Received 27 September 1968)

The effect on the isotropic spin density of the $(\mathbf{k},\mathbf{k}')$ dependence of the exchange coupling $J(\mathbf{k},\mathbf{k}')$ between a localized magnetic moment and conduction electrons predicted by Ruderman-Kittel-Kasuya-Yosida (RKKY) theory is determined quantitatively. Spherical local moments are employed, viz., Gd$(4f^7)$ and Fe$(3d^5)$ (which are taken as representative of rare-earth and transition metal moments, respectively). The conduction bands are described by simple orthogonalized plane waves appropriate to a "free-electron" metal with $\mathbf{k}$ the wave vector of the incident electron and $\mathbf{k}'$ that of the scattered electron. We find that a Q-dependent coupling (where $Q\equiv|\mathbf{k}-\mathbf{k}'|$) has some justification when dealing with a Gd local moment but has considerably less justification for Fe. Both the $(\mathbf{k},\mathbf{k}')$ and the Q-coupling schemes yield a "main" spin-density peak which is more diffuse than that yielded by coupling approximations traditionally applied to RKKY theory. Spin-density results were obtained which are appropriate to the outer reaches of a lattice site and to the nuclear site of either the local moment or neighboring atoms (these involve inclusion of core s terms in the spin density). These results suggest that spin distributions obtained by neutron diffraction and those inferred from hyperfine field measurements may differ significantly.

I. INTRODUCTION

THE response of conduction electrons to the exchange interaction with localized impurity spins appears to play an increasingly important role in the explanation of various phenomena observed in metals and dilute alloys. This so-called s-d exchange interaction has been the object of extensive investigations, ranging from the well-known case of dilute alloys such as Pd-Fe to the variety of observations (resistance minima, NMR, Mössbauer effect, susceptibility, and specific heat) attributed to the existence of a Kondo bound state. The basic starting point often taken for more detailed theoretical treatments is the familiar Ruderman-Kittel-Kasuya-Yosida (RKKY) model.[1] The s-d exchange-coupling integral $J(\mathbf{k},\mathbf{k}')$ between the localized magnetic electrons and conduction electrons is commonly assumed to be constant, or at best only a function of the magnitude of the wave-vector difference $Q=|\mathbf{k}'-\mathbf{k}|$ between incident and scattered conduction electrons. In low order, this assumption leads to an induced conduction-electron spin density whose Fourier transform $\rho(Q)$ is simply related to $\chi(Q)$, the zero-frequency magnetic susceptibility of the conduction electrons, by $J(Q)$, i.e.,

$$\rho(Q)\approx J(Q)\chi(Q). \tag{1}$$

(Similar assumptions are made concerning the Coulomb terms entering some of the more advanced theories.)

We have previously described investigations[2] (henceforth referred to as I) of the behavior of the exchange integral $J(Q)$ over a range of Q values for the illustrative case of a local moment which consists of the half-filled shell of $Gd^{3+}(4f^7)$ and conduction electrons which are treated as orthogonalized plane waves (OPW's); comparison was made with the traditional assumptions about the nature of $J(Q)$ as used in RKKY theory. The resulting spin density of the conduction electrons, with and without the exchange enhancement[3] of the susceptibility $\chi(Q)$, was found to be severely affected by the assumed form of $J(Q)$. Use of the actual computed exchange coupling was shown to appreciably reduce qualitative disagreement between theory and experiment. As we found,[2] it is only by numerical *accident* that a local-moment–conduction-electron coupling can be qualitatively described by a Q-dependent J. Assuming that $J(\mathbf{k},\mathbf{k}')=J(Q)$ allows one to apply knowledge of the magnetic susceptibility to the problem(s) at hand, but unfortunately the *actual response* of the metal to the local moment is more complicated. Exchange coupling is, after all, a nonlocal interaction.

In this paper, we inspect the effect of the actual $(\mathbf{k},\mathbf{k}')$ dependence of $J(\mathbf{k},\mathbf{k}')$ on the spin density predicted by RKKY theory. (In our view, the quantitative changes so introduced are as important as some of the higher-order refinements of expressions, such as Eq. (1), which have been introduced so as to bring theory and experiment into better agreement.) Following the original development of RKKY theory, we deal with the bands of a "free-electron" metal which are de-

* Work performed under the auspices of the U. S. Atomic Energy Commission.

† Supported in part by the Advanced Research Projects Agency at the Northwestern Materials Research Center.

[1] M. A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954); T. Kasuya, Progr. Theoret. Phys. (Kyoto) **16**, 45 (1956); K. Yosida, Phys. Rev. **106**, 893 (1957); S. Vonsovski, Zh. Eksperim. i Teor. Fiz. **16**, 981 (1946), **24**, 419 (1953); A. H. Mitchell, Phys. Rev. **105**, 1439 (1957).

[2] R. E. Watson and A. J. Freeman, Phys. Rev. **152**, 566 (1966), henceforth denoted as I.

[3] For example, P. A. Wolff, Phys. Rev. **120**, 814 (1960); **129**, 84 (1963); B. Giovannini, M. Peter, and J. R. Schrieffer, Phys. Rev. Letters **12**, 736 (1964).

scribable in terms of plane waves or simple OPW's. In this case, the coupling associated with a spherical local moment takes the obvious form

$$J(\mathbf{k},\mathbf{k}')=\sum_L P_L(\cos\Omega)F_L(k,k'), \tag{2}$$

where Ω is the angle between $\mathbf{k}$ and $\mathbf{k}'$ and F is a function of only the magnitudes of $\mathbf{k}$ and $\mathbf{k}'$. Equation (2) is not readily approximated by a $J(Q)$. Unfortunately, a theory acknowledging this $(\mathbf{k},\mathbf{k}')$ coupling is both computationally unpleasant and yields results which are in a much less convenient form than Eq. (1). Despite this, $(\mathbf{k},\mathbf{k}')$ results are of interest in their own right and for two other reasons as well.

First, many experiments (most notably hyperfine interaction measurements) sample the conduction-electron spin density in the interior of an ion core where the core character assumed by the conduction electrons affects the measured spin-density distribution. This is rigorously dealt with in a $(\mathbf{k},\mathbf{k}')$ treatment[4] of the density and results appropriate to the Fermi contact interaction at nuclear sites will be obtained in this paper. These will be compared with simple plane-wave density predictions which are relevant to the full spin-density behavior in the outer reaches of a Wigner-Seitz cell. The results suggest that spin distributions obtained by neutron diffraction and those inferred from hyperfine field measurements may differ significantly.

Second, the *effective* exchange coupling, associated with interband mixing,[5,6] assumes roughly[6] the form of a *single* $L\neq 0$ term of the type seen in Eq. (2), in which case Eq. (1) becomes entirely inappropriate. Such interband mixing will be inspected in a subsequent paper; the computational machinery necessary for this is identical to that employed in the present paper.

We here limit consideration to spherical local moments and results are reported for the polarization induced by a Gd($4f^7$) and an Fe($3d^5$) local moment. These results are assumed to be typical of the isotropic polarization appropriate to the rare earths and to the iron series elements, the moment being in the interior of the "ion core" in the former case and in the "exterior" in the latter. Many features of the results of I for Gd, where $J(\mathbf{k},\mathbf{k}')$ was approximated by a $J(Q)$, which was then employed in Eq. (1), will be seen in the more exact estimates of $J(\mathbf{k},\mathbf{k}')$ obtained here. The same $J(Q)$ approximation will be shown to be less justified for Fe-moment–conduction-electron coupling.

[4] For a spherical local moment these core terms can be incorporated into the F_L of Eq. (2). Kaplan has attempted [Phys. Rev. Letters **14**, 499 (1965)] to incorporate them into the $\chi(Q)$ of Eq. (1). At best, this can be only crudely done and our results differ from his. Some reasons for this will be obvious in Sec. II.

[5] P. W. Anderson and A. M. Clogston, Bull. Am. Phys. Soc. **2**, 124 (1961); J. Kondo, Progr. Theoret. Phys. (Kyoto) **28**, 846 (1962); S. Koide and M. Peter, Rev. Mod. Phys. **36**, 160 (1964).

[6] R. E. Watson, S. Koide, M. Peter, and A. J. Freeman, Phys. Rev. **139**, A167 (1965).

II. THE MODEL

In traditional RKKY theory, the spin density arises from the spin repopulation associated with the Pauli susceptibility and from the lowest-order perturbation mixing of unoccupied Bloch orbital character $\varphi_{\mathbf{k}'}$ into the occupied Bloch states $\varphi_{\mathbf{k}}$, i.e.,

$$\varphi_{\mathbf{k}\pm}\rightarrow\varphi_{\mathbf{k}\pm}\mp S\sum_{k'>k_F}\frac{J(\mathbf{k},\mathbf{k}')}{\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}}\varphi_{\mathbf{k}'\pm}, \tag{3}$$

where k_F is the Fermi wave vector, the $\epsilon_{\mathbf{k}}$'s are conduction-electron orbital energies, the $\pm$ subscripts (and the $\mp$ of the sum) refer to spin parallel and antiparallel to the local moment of spin S, and

$$J(\mathbf{k},\mathbf{k}')\equiv\left\langle\int\int d\tau_1 d\tau_2\varphi_{\mathbf{k}}^*(\mathbf{r}_1)\psi_i(\mathbf{r}_1)\times\frac{1}{r_{12}}\varphi_{\mathbf{k}'}(\mathbf{r}_2)\psi_i^*(\mathbf{r}_2)\right\rangle_i, \tag{4}$$

where the average is over the unpaired local-moment orbitals ψ_i. For a spherical moment and simple OPW's, $J(\mathbf{k},\mathbf{k}')$ has the form of Eq. (2). Keeping contributions linear in J, the Pauli and mixing terms together yield the familiar expression

$$\rho_\pm(r)=\mp S\sum_{k=0}^{k_F}\sum_{k'=0}^{\infty}\frac{J(\mathbf{k},\mathbf{k})}{\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}}\times[\varphi_k^*(\mathbf{r})\varphi_{k'}(\mathbf{r})+\varphi_k(\mathbf{r})\varphi_{k'}^*(\mathbf{r})], \tag{5}$$

where $\pm$ again refers to majority and minority spins and the odd symmetry (in k and k') of the energy denominator has been used to extend the k' summation over all k space.[1]

The total spin density is

$$\rho(r)=\rho_+(r)-\rho_-(r). \tag{6}$$

Assuming J to be a function of Q and substituting plane waves for the φ, one may obtain Eq. (1), apart from numerical constants.

Neither the assumption of a $J(Q)$ nor a plane-wave φ_k is appropriate to the problem at hand. Central to both these matters is the form of φ_k appropriate to a "free-electron" band in a metal with ion cores present, namely, a single, or linear combination, of OPW's. A single OPW, when at site $\mathbf{R}$ in the lattice, has the form

$$\varphi_k(\mathbf{r})\equiv\frac{1}{\sqrt{V}}N_k W_k(\mathbf{r}-\mathbf{R})e^{i\mathbf{k}\cdot\mathbf{R}}=\frac{1}{\sqrt{V}}N_k e^{i\mathbf{k}\cdot\mathbf{R}}\sum_{lm}4\pi(i)^l\Xi_{l,m^k}(|\mathbf{r}-\mathbf{R}|)\times Y_l^m(\theta,\varphi)Y_l^{-m}(\theta_k,\varphi_k), \tag{7}$$

where V is the atomic volume associated with a lattice site (to which we will normalize results). Such a single OPW will be employed throughout this paper. For a spherical core, Ξ is independent of m and is of the form

$$\Xi_l^k(r)=j_l(kr)-\sum_{n,l} S_{nlk}R_{nl}(r), \tag{8}$$

where j_l is a spherical Bessel function, $R_{nl}(r)$ is the radial function of the nl core shell, and

$$S_{nlk}\equiv\int j_l(kr)R_{nl}(r)r^2dr \tag{9}$$

is the overlap between the radial part of the plane wave and the core radial function. The OPW normalization factor (again for spherical cores) is given by

$$N_k=\left\{1-\frac{4\pi}{V}\sum_{nl}(2l+1)S_{nlk}^2\right\}^{-1/2}, \tag{10}$$

the volume normalization having already been incorporated into (7). For spherical cores, the $W_\mathbf{k}$, $\Xi_\mathbf{k}$, and $N_\mathbf{k}$ are functions only of the magnitude of $\mathbf{k}$ which is a great computational convenience.

Insertion of the φ into Eq. (4), for an assumed spherical moment, readily shows that the Lth components Ξ_L^k and $\Xi_L^{k'}$ of the OPW's are associated with the Lth term in the exchange coupling [Eq. (2)]. In turn, inserting Eq. (2) into the density expression, carrying out the angular integrations and evaluating the density at a nuclear site $r=R$, one obtains, apart from constants,

$$\rho_\pm(r)\sim\mp S\sum_L\int\int k^2dkk'^2dk'\left\{\frac{F_L(k,k')}{k^2-k'^2}j_L(kR)\times j_L(k'R)W_k(0)W_{k'}(0)N_kN_{k'}\right\}. \tag{11}$$

Therefore, only the $L=0$ term contributes to the spin density at the nucleus at the local moment's site ($\mathbf{R}=0$). This implies that interband mixing will not significantly affect the Fermi contact interaction at this site since effective interband exchange coupling involves a single $L\neq0$ term in low order.[6] The various L partial waves of the disturbance are driven by the exchange coupling involving OPW components of the same L. Once away from the origin, all partial waves contribute to the contact interaction.

Now the Pauli exclusion principle, as expressed in the antisymmetry of a Hartree-Fock function, requires either that the conduction electron φ be orthogonal to the occupied core states or that their nonorthogonality be explicitly accounted for when the expectation values of physical quantities such as J are evaluated. Both lead to the same quantitative predictions. In other words, the employment of OPW's yields *the* correct results for a "free-electron" metal with cores present.

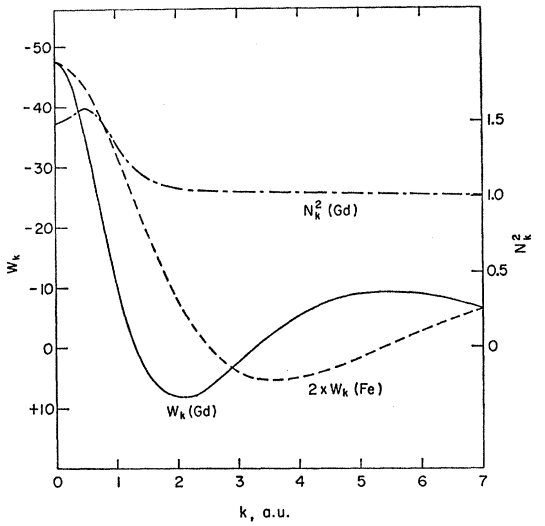

FIG. 1. The nuclear-site core-density factors $W_k(0)$ for Gd and Fe [see Eq. (12)]. Also, the core renormalization factor N_k^2 for Gd.

The use of OPW's can be justified in a second way: OPW's supply a basis for constructing conduction-electron eigenfunctions of the crystal potential (this point of view was important to the development of OPW energy band theory). Given these observations it is quite clear that the sums of Eqs. (7), (8), and (10) should span the closed shells of the ion cores with appropriate n and l when one in turn evaluates equations such as (4) and (5). It is less clear what should be done to account for the Pauli principle in the case of only partially occupied shells like the $3d$ and $4f$ in Fe and Gd, respectively.

The OPW φ-Gd($4f^7$) exchange coupling was described in detail previously in I. $J(\mathbf{k},\mathbf{k}')$ consists almost entirely of $L=0$ and 1 terms [see Eq. (2)] which combine in such a way as to be well approximated by a $J(Q)$. Orthogonalization to the closed $5s$ and $5p$ shells of the ion (which lie exterior to the open $4f$ shell) strongly affected the quantitative character of $J(\mathbf{k},\mathbf{k}')$ and was essential[7] to the success of the $J(Q)$ fit. A similar fit does not work for the coupling of an Fe d^5 moment. Here, $L=0$, 1, and (to a lesser degree) 2 terms[8] are significant.

The φ explicitly entering the spin-density expression [Eq. (5)] are also affected by core character. Let us consider two cases: (1) the density for an interstitial

[7] It should be noted that a $J(\mathbf{k},\mathbf{k}')$ calculated for Gd in the incorrect way using plane wave φ's is not well approximated by a $J(Q)$ (see I).

[8] The $L=2$ term for Fe was evaluated including orthogonalization to all orbitals of the open $3d$ shell. Such a treatment is convenient but not rigorously correct since orthogonalization is properly done to occupied orbitals alone. Perhaps the best choice of treatment is to simultaneously treat the $3d$-plane-wave (interband) mixing; we will not do this here. Our choice of orthogonalization does not affect the qualitative character of our results. The $L=3$ term for Gd similarly includes orthogonalization with all orbitals of the open $4f$ shell. This has no visible effect on the Gd results.

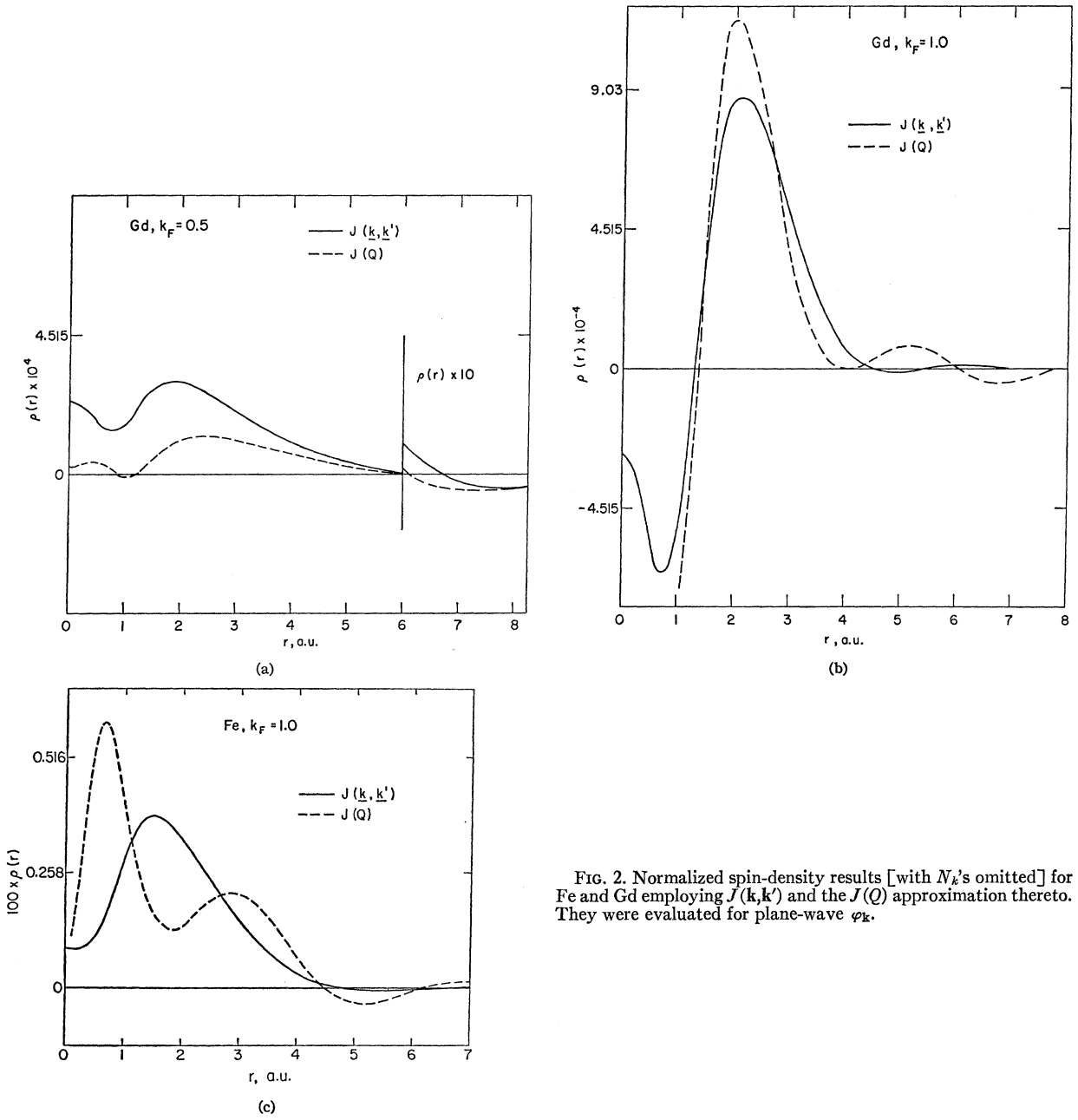

FIG. 2. Normalized spin-density results [with N_k's omitted] for Fe and Gd employing $J(\mathbf{k},\mathbf{k}')$ and the $J(Q)$ approximation thereto. They were evaluated for plane-wave $\varphi_{\mathbf{k}}$.

position where the traditional plane-wave approximation [i.e., $W_k(\mathbf{r}-\mathbf{R})=e^{i\mathbf{k}(\mathbf{r}-\mathbf{R})}$] should apply, and (2) the density at a nucleus for which

$$W_{\mathbf{k}}(0)=[1-\sum_n S_{n0k}R_{n0}(0)], \qquad (12)$$

where the sum now spans core s shells.

The $W_{\mathbf{k}}(0)$ for Gd and Fe, which are plotted in Fig. 1, are seen to oscillate.[9] It is quite clear, since they are functions only of the magnitude of $\mathbf{k}$, that a product $W_{\mathbf{k}}\cdot W_{\mathbf{k}+\mathbf{Q}}$ cannot be well represented by a Q-dependent function [i.e., this core character cannot be readily inserted into Eq. (1)]. Note that the spin density may be severely affected if a node in W_k falls near k_F—a possibility for Gd, where the first node occurs at ~1.3 a.u. The W_k nodes for Gd occur at smaller k since it has a spatially larger s core than Fe. The square of the normalization constant N_k^2 for Gd (evaluated with

[9] These, the N_k^2 of Fig. 1 and the exchange integrals were evaluated with free Fe^{2+} [R. E. Watson, Technical Report No. 12, MIT, 1959 (unpublished)] and Gd^{3+} [A. J. Freeman and R. E. Watson, Phys. Rev. **127**, 2058 (1962)] ion wave functions.

an atomic volume appropriate to Gd metal) is also plotted. It falls off slowly, has little structure, and, as will be seen, affects little but the normalization of the spin-density response.

III. SPIN DENSITIES: COMPARISON BETWEEN $J(Q)$ AND $J(k,k')$ RESULTS

Spin-density results employing computed $J(\mathbf{k},\mathbf{k}')$ and $J(Q)$ as estimated (see I) therefrom are plotted in Fig. 2 for plane wave φ_k. For Gd, plots are shown for $k_F=0.5$ and 1.0 a.u. and for Fe for $k_F=1.0$ a.u. Note that a typical "free-electron" metal has k_F lying between 0.5 and 1.0 a.u. The Gd $J(Q)$ densities are those already reported in I; the Fe $J(Q)$ and the associated density results are from new calculations obtained in the same way. The $J(\mathbf{k},\mathbf{k}')$ results were obtained by numerical integration of Eq. (5) omitting the k dependence of the N_k's. The singularity in the integrand (where $k\rightarrow\mathbf{k}_F$, $k'\rightarrow\mathbf{k}_F$) was dealt with analytically. Although the densities are seen to oscillate, we are well inside the asymptotic region at the largest r shown.[10] These results[9,11] are for normalized φ_k, except for the omission of the N_k's.

Since both the $J(Q)$ and $J(\mathbf{k},\mathbf{k}')$ estimates treat the Pauli or $J(0)$ term in the same way, the predicted total spin density is identical for both. As expected, the $k_F=0.5$ a.u. results for Gd show the best agreement between densities since it appears[2] that the $J(Q)$ approximation is best here. The strongest differences occur for the Fe results; in particular, a change in the structure of the density curve occurs at small r. Results for Fe at smaller k_F (not plotted here) show better agreement. Characteristic oscillations, with periods $\sim 1/k_F$, occur at the large (but still small) r values for all results (this behavior is typical though we are not[10] in the asymptotic, i.e., very large r, region for which the oscillations are normally derived). In all cases the $J(Q)$ and $J(\mathbf{k},\mathbf{k}')$ results differ as to the phase of the oscillations. For $k_F=1$, the $J(Q)$ predictions, for either local moment, overestimate the amplitude of these oscillations by an order of magnitude (while maintaining the correct total spin). Discrepancies in phase and amplitude of this sort have severe implications for any quantitative estimate involving a sum or scan over lattice sites such as one does, for example, when evaluating "wipe out" numbers or solvent Knight shifts appropriate to NMR.

[10] That is, the immediate k-space region about the singularity in the integrand of Eq. (5) plays but a minor role in the results. The traditional asymptotic analytic derivation of the oscillations is appropriate to radial distances many times those plotted here.

[11] Significant differencing occurs in the course of integration for all but the smallest r values. Due to this, some uncertainty must be attached to the results for the larger r shown. We believe that large r density amplitudes and nodes to be better determined than is required for purposes here. However, they are not accurate enough to determine a power law for the amplitude of the outer oscillations.

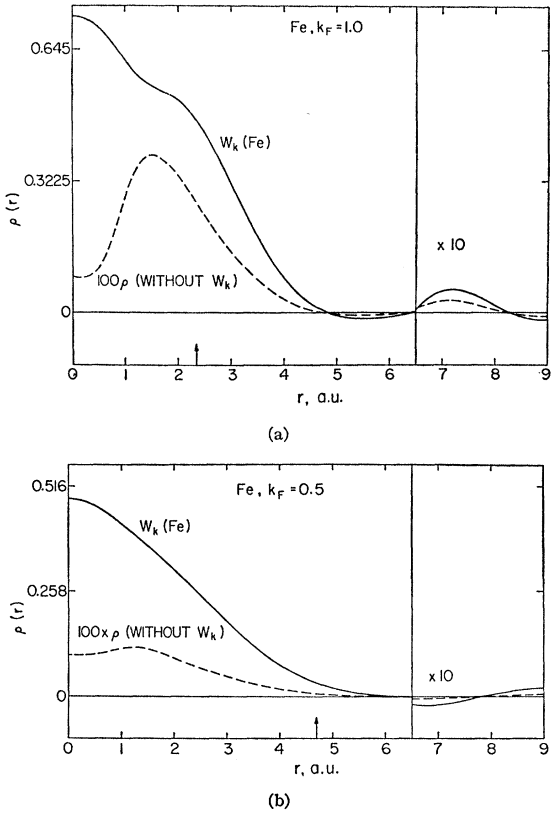

FIG. 3. Normalized Fe spin-density results, labeled "without W_k" for interstitial regions (i.e., for plane-wave φ_k). N_k renormalization factors are omitted.

IV. CORE EFFECTS ON SPIN DENSITIES

Spin-density results employing the plane wave $W_k(\mathbf{r}-\mathbf{R})$ and the $W_k(0)$ appear for Fe in Fig. 3 (N_k factors have again been omitted). Note that the contact, or $W_k(0)$, density plots imply the presence of an Fe (or similar $3d$) atom centered with its nucleus at the r value in question. This is clearly physically impossible, except at the origin, until one is out at some distance corresponding to a typical nearest-neighbor lattice spacing, i.e., 4 or 5 a.u. Results are nevertheless plotted at smaller r because they are indicative of how the actual density differs from the plane-wave prediction due to the presence of core terms in the φ_k. The curves differ in shape out to a distance of 5 or 6 a.u. Outside this region they oscillate, differing only in amplitude. The ratios of the $W_k(0)$ to the plane-wave density amplitude are quite accurately given by the square of $W_{k_F}(0)$ (i.e., evaluated at k_F) as might be expected. The results at the origin, which correspond to the Fermi contact density at the local moment's own nucleus, are considerably enhanced when the core terms are accounted for.

Similar behavior, with one important exception, is seen for the Gd results plotted in Fig. 4. Here the contact density induced by the Gd moment at another

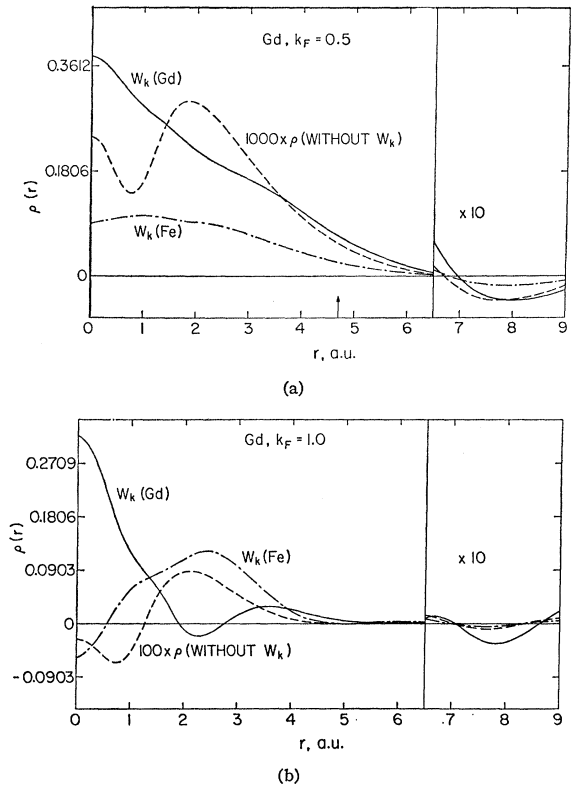

FIG. 4. Normalized Gd spin-density results, labeled "without W_k" for interstitial regions (i.e., for plane-wave φ_k) and for nuclear sites [employing the $W_k(0)$]. N_k renormalization factors omitted.

nuclear site a distance r away is obtained for either a Gd or an Fe atom at that site. These may be taken as typical of the response to the polarization induced at neighboring lanthanide and $3d$ atom sites by a Gd impurity. We again see variation in the shape of the distributions out to 5 or 6 a.u., and amplitudes which scale roughly as $[W_{k_F}(0)]^2$, but now there also are small variations in the phase of the outer oscillations. The more pronounced changes, seen here, are associated with the more rapid fall off of the $W_k(0)$ for Gd seen in Fig. 1.

The spin distributions seen in Figs. 3 and 4 are more diffuse than their traditional RKKY counterparts. The first node of the traditional RKKY function is indicated by an arrow on the plots and, as a general rule, this falls well inside where the present spin distributions node. Whereas the traditional results almost always go negative before near-neighbor nuclear sites are reached, the present results indicate that near neighbors may lie inside the "main peak" of the distribution. It should be further noted that the atomic $4f$ and $3d$ spin densities have gone almost to zero at a radius of 2.5 to 3 a.u.; thus, the induced spin density, residing in the main peak of the density distribution, may be significantly more diffuse than the local moment which causes it, giving it the potential of being seen experimentally.

The conduction-electron spin density at the local moment nucleus has, in each case, become large and positive when core s terms were accounted for. While it is possible that it is negative for some special case, it appears that the possibility of very small or negative contact densities (i.e., antiparallel to the moment spin direction) are much less likely than we suggested in I. We might note that the ratios of the densities at the origin to those appropriate to near-neighbor distances are still much smaller than those predicted by traditional theory. In other words, a substantial hyperfine field induced, and seen, at a neighboring ion site need not imply one several orders of magnitude larger at the nucleus of the local moment.

Whether at a local moment site or in the neighboring region, core s terms cause the spin density at a nuclear site to be considerably larger than that appropriate to plane waves (which presumably has some relevance to the spin behavior in regions between atoms in the crystal). This enhancement is crudely given by $[W_{k_F}(0)]^2$. Now it is frequent practice to take an experimental hyperfine field, obtain a contact density from it, and in turn infer the strength of the local-moment–conduction-electron coupling. It is clear, however, that the omission of this enhancement factor (plus any ill assumptions concerning the shape of the spin distribution) can yield misleading estimates for this coupling.

V. OPW NORMALIZATION EFFECTS

All the results reported have been obtained omitting the N_k which account for the additional normalization due to core orthogonalization [cf. Eq. (10)]. Results with and without these terms appear in Fig. 5 and were obtained for Gd with an atomic volume appropriate to Gd metal. The spin density has simply been scaled by roughly a factor of $N_{k_F}{}^4$ (the fourth power occurs since the normalization appears in both the exchange integral and the spin density). Otherwise, the density distribution has been only slightly distorted by the weak effects associated with the small variation seen for N_k in Fig. 1. As a general rule, little more than a scaling should be expected from this normalization factor. Exceptions might occur for cases where an atom is inserted into a lattice volume which is small compared with ion core "size" for then N_k would vary more violently [again see Eq. (10)].

VI. CONCLUSION

We have concentrated in this paper on presenting spin-density results obtained with the actual $J(\mathbf{k},\mathbf{k}')$ local-moment–conduction-electron exchange integral appropriate to the illustrative cases of Gd and Fe moments. We have not made detailed comparison with experiment. There are several obvious reasons for this which should be mentioned at this time.

Pure metals, or intermetallic compounds whose constituent atoms have local moments, are, in general, not describable in terms of free-electron bands. This is obviously true for transition metals, but holds also for the rare earths which are, in fact, transition metals too.[12] As a result, the model we employ can make little contact with experimental reality for these systems.[13] One might hope that the present model and, hence, our results are relevant to the case of a local moment as an impurity in a free-electron metal. Unfortunately, there are difficulties here too. For example, the local-moment atom will amost inevitably be a charge impurity and the resulting charge screening should affect the spin-density response. Also, the φ_k, for the random alloy, will not form an orthogonal basis set and additional terms should be included in the spin-density description. The difficulty in relating free-electron predictions to experiment is thus considerable. We have considered the model here because, with rare exception, it is this model (or slight variations thereof) which is employed and compared with experiment. Insight into what the model *actually* predicts in low order is thus useful in itself and a necessary first step before asking what actually happens quantitatively in these metals.

Despite the difficulties, there are a number of qualitative features of our results which are relevant to experiment. We see that the bulk spin density as measured by neutron magnetic scattering need not be faithfully reflected in the density distribution seen by nuclei in a hyperfine measurement. Correlation of the two may thus be difficult. The presence of core effects in metals is widely recognized elsewhere, but somehow it has been almost overlooked[4] and its implications ignored for the *s-d* interaction. As in I, we see the range

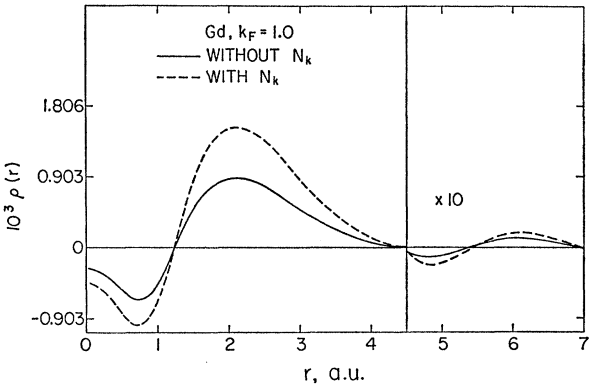

FIG. 5. Normalized Gd spin-density results obtained with and without the N_k core renormalization factors and for plane-wave φ_k.

of the "main peak" of the induced spin distribution to be significantly greater than that yielded by more traditional estimates of the RKKY model. Near-neighbor sites are, more likely than not, to lie within the mean peak. Exchange enhancement of the response,[3] of course, further increases this "range." In addition, we have seen that the amplitude and phase of the near oscillations are sensitive to details of the coupling. Lower amplitudes are obtained with $J(\mathbf{k},\mathbf{k}')$ than with the $J(Q)$ approximation. This result and questions regarding the range of the main peak would suggest that the frequent usage of asymptotic density expressions in what are near neighbor regions is at best doubtful and at worst simply wrong. Hence, much more needs to be known of spin-density distributions before one can safely infer such quantities as exchange constants from experiment.

ACKNOWLEDGMENTS

These results were obtained with the invaluable programming assistance of B. Cetlin and E. Wolfson. One of us (AJF) thanks T. G. Bear for helpful discussions.

[12] For example, J. O. Dimmock and A. J. Freeman, Phys. Rev. Letters 13, 750 (1964).

[13] Results have been obtained, and further work is underway to obtain the susceptibility for the "real" bands of transition metals at a number of institutions [e.g., G. Allan, W. M. Lomer, R. D. Lowde, and C. G. Windsor, Phys. Rev. Letters 17, 933 (1968)]. These efforts will eventually involve considerations similar to, but different from those described here.