Self-consistent screening of a positive muon in a spin-polarized electron gas*

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The electron spin-density distribution around a positive muon has been calculated self-consistently for a range of metallic densities using the spin-density functional formalism. The enhancement of the spin density at the μ^+ site over the ambient polarization is found to be considerably smaller than the corresponding charge-density enhancement. The results are used to estimate the hyperfine field at interstitial positive-muon sites in ferromagnetic Fe, Co, Ni, and Gd. Comparison is made with the most-recent experimental data.

I. INTRODUCTION

The distribution of internal fields in ferromagnetically ordered systems has been a topic of interest. Fourier transform of the neutron scattering form factors can reveal the spatial dependence of magnetization density of magnetic materials. The local change of this magnetization density due to an impurity (substitutional or interstitial) can be probed by a variety of experimental techniques through hyperfine-field measurements. Most of the impurities have atomic character and an analysis of the spin density at such impurity sites is often hindered by the presence of core electrons. However, this difficulty does not arise for an impurity such as a positive muon.

A positive muon carries a unit positive charge with a mass ~200 times the electron mass. Like protons, positive muons are believed to occupy interstitial sites in a metal. The internal field B, experienced by a muon, is measured through its spin rotation[1] and is the sum of three contributions:

$$B = \tfrac{4}{3}\pi M + B_d + B_{hf} \ , \tag{1}$$

where B_d is the field due to the local dipole moments inside a sphere centered on the muon site and $\frac{4}{3}\pi M$ is the so-called Lorentz field due to induced magnetic charges on the surface of the sphere. B_d is obtained from a knowledge of the crystal geometry of the host metal. Thus, knowing the sign and magnitude of B, one can extract the contribution to the internal field due to the hyperfine coupling B_{hf} of the muon spin with the spin-polarized conduction band.

The hyperfine field at a μ^+ site in a ferromagnetic material can be written

$$B_{hf} = -\tfrac{8}{3}\pi\mu_B[n^+(0) - n^-(0)] \ , \tag{2}$$

where $n^+(0)$ and $n^-(0)$ are, respectively, the densities of electrons with spin ↑ and spin ↓ at the muon site. The sign convention used here is such that the spin-up (↑) electron has a magnetic moment of $-\mu_B$ and hyperfine fields are positive when parallel to the magnetization in the positive z direction. A positive muon attracts electrons of both spin, and as a result, the electron charge density at the muon site $n(0) = n^+(0) + n^-(0)$ and the spin density $P(0) = n^+(0) - n^-(0)$ will be considerably enhanced over the background charge density n_0 and spin density, respectively. Since the positive muon represents a strong perturbation, the calculation of either the charge or spin polarization is a nonlinear one. Hence, the problem of the spin density enhancement over the ambient polarization as a function of electron density $n_0 = 1/\frac{4}{3}\pi(r_s a_0)^3$ (r_s conventionally being referred to as the electron density parameter) is interesting. One might naively imagine that the electron density per spin would be enhanced in the same proportion as the charge density. Jena[2] has treated this problem by ap-

proximating the effective muon-electron potential by a spin-dependent square well. Patterson and Falicov,[3] within a linear-response calculation, have shown that the spin-density enhancement

$$[n^+(0)-n^-(0)]/(n_{+0}-n_{-0})$$

is much less than the corresponding charge-density enhancement for r_s appropriate to ferromagnetic Ni. Petzinger and Munjal[4] have treated the same problem within the spin-density functional formalism[5,6] and obtained approximately self-consistent solutions for a μ^+ in Ni. While their result[4] for the individual charge and spin density enhancements differs considerably from that of Patterson and Falicov,[3] they also find that the spin density is enhanced to a much smaller degree than the charge density.

The purpose of this paper is to make a careful investigation of the relative enhancements of charge and spin densities as a function of the bulk electron density and ambient spin polarization. We have calculated self-consistently the charge and spin density distribution around a static μ^+ using the spin-density functional formalism for densities $1 \leq r_s \leq 5$. In Sec. II, we briefly review the spin-density formalism and outline our numerical procedure. Our results are presented in Sec. III. We make use of these results to estimate the hyperfine field at a μ^+ site in ferromagnetic Fe, Co, Ni, and Gd in Sec. IV.

II. SPIN-DENSITY FUNCTIONAL FORMALISM

In the case of a strongly polarized electron gas, the set of Hohenberg-Kohn-Sham (HKS) equations[5,6] which determine the spin density $n^\sigma(r)$ is

$$[-\nabla^2+V^\sigma_{\rm eff}(\vec{r})]\psi_i^\sigma(\vec{r})=\epsilon_i^\sigma\psi_i^\sigma(\vec{r}) \quad . \tag{3}$$

We have used atomic units (energy in Rydbergs and length in Bohr radius) throughout this paper. The density of electrons per spin σ is given by

$$n^\sigma(\vec{r})=\sum_i^{\rm occ}|\psi_i^\sigma(\vec{r})|^2 \quad . \tag{4}$$

The effective potential in Eq. (3) is obtained from the exact ground-state energy density E through

$$V^\sigma_{\rm eff}(\vec{r})=\delta E/\delta n^\sigma(\vec{r}) \quad . \tag{5}$$

For a spherically symmetric potential, Eq. (3) becomes

$$\left[-\frac{d^2}{dr^2}+\frac{l(l+1)}{r^2}+V^\sigma_{\rm eff}(r)\right]R^\sigma_{kl}(r)=(k^\sigma)^2R^\sigma_{kl}(r) \quad , \tag{6}$$

where $R^\sigma_{kl}(r)$ is the radial part of the wave function $\psi^\sigma_{kl}(\vec{r})$ and k^σ is the wave vector for an electron with spin σ. The effective potential, in the local density approximation, in Eq. (6) is

$$V^\sigma_{\rm eff}(r)=\frac{-2}{r}+2\int d^3r'\,\frac{n(\vec{r}')-n_0}{|\vec{r}-\vec{r}'|}+\mu^\sigma_{xc}(n,\zeta)-\mu^\sigma_{xc}(n_0,\zeta_0) \quad . \tag{7}$$

The first two terms in the above equation are the external and Hartree potentials, respectively. The exchange-correlation potential, μ^σ_{xc} in the local-density approximation depends on the charge density $n(r)=n^+(r)+n^-(r)$ and the spin density $\zeta(r)=[n^+(r)-n^-(r)]/n(r)$ and has been taken from the work of Gunarsson *et al.*[7] The electrostatic term goes to zero at large r because of charge screening. For the exchange-correlation potential to be zero at large distances, we subtract a uniform contribution, $\mu^\sigma_{xc}(n_0,\zeta_0)$ that corresponds to the background charge density n_0 and spin density $\zeta_0=(n_{+0}-n_{-0})/n_0$. The polarization can be thought of as being maintained by a stabilizing external magnetic field H_0, the strength of which can be determined from the minimal condition of total free energy. For a sufficiently strong potential $V^\sigma_{\rm eff}$, bound state solutions exist for Eq. (6) with $k^{\sigma 2}\rightarrow -E_b^\sigma$, the bound-state energy below the band continuum. Thus, including bound states the deviation of the electron density per spin from its ambient value is given by

$$\delta n^\sigma(r)=n^\sigma(r)-n_0=\frac{1}{2\pi^2}\int_0^{k_F^\sigma}dk\,k^2\sum_l(2l+1)\times\left(\frac{[R^\sigma_{kl}(r)]^2}{r^2}-j_l^2(kr)\right)+\left(\frac{R_b^\sigma(r)}{r}\right)^2 \quad , \tag{8}$$

where $R_b^\sigma(r)$ is the radial part of the bound-state wave function in the $l=0$ state, and k_F^σ is the Fermi wave vector for spin σ,

$$k_F^\pm=(1\pm\zeta_0)^{1/3}(\tfrac{9}{4}\pi)^{1/3}(1/r_s) \quad . \tag{9}$$

The bound-state wave functions for each spin σ are normalized to unity; and the scattering-state wave functions are matched to the asymptotic solution at large r,

$$R^\sigma_{kl}(r)\underset{r\rightarrow\infty}{=}\cos\delta_l^\sigma(\epsilon_k)\,j_l(kr)-\sin\delta_l^\sigma(\epsilon_k)\,n_l(kr) \quad , \tag{10}$$

where δ_l^σ's are the scattering phase shifts for lth partial wave and j_l and n_l are, respectively, spherical Bessel and Neumann functions of order l. The scattering phase shifts at the Fermi energy satisfy the sum rule

$$Z^{\sigma}=\frac{1}{\pi}\sum_{l}(2l+1)\delta_l^{\sigma}(\epsilon_F)\ ,$$
$$Z=1=Z^{+}+Z^{-}\ . \tag{11}$$

Equations (6)–(8) are solved self-consistently for different values of the parameters r_s and ζ_0. Since the exchange-correlation potential for spin ↑ depends also on the population of spin ↓ and vice versa, the problem reduces to solving two sets of coupled Schrödinger-like equations self-consistently.

Once the exchange-correlation potential per spin is prescribed, the above self-consistent procedure appears to be simple and straightforward. In practice, however, the system of Eqs. (6)–(8) does not yield convergent solutions. As in the case of screening of a proton in paramagnetic electron gas, the difficulty lies in the long-range nature of the Coulomb potential. Due to numerical inaccuracies, the sum rule in Eq. (11) in a given iteration may differ slightly from unity. As a result the electrostatic potential in Eq. (7) has a long range which causes the subsequent iterations to diverge. In a recent calculation, Petzinger and Munjal[4] have overcome this difficulty by parametrizing the effective potential. Thus, they were able to obtain approximately self-consistent solutions for spin densities for $r_s=3.2$. We have instead followed the procedure of Manninen *et al.*[8] as described in one of our earlier papers.[9] This involves no parametrization of the effective potential and always leads to a fully convergent solution within about 20 iterations. Even at the origin, our spin densities are accurate to better than 5%, with rapidly increasing stability as r increases.

We have solved Eq. (6) by dividing the interval between 0 to k_F^{σ} in twenty steps for each value of l, $0\leq l\leq 10$. For the variable r, we have considered a mesh of 200 points in steps of $0.1a_0$. The effective potential in Eq. (7) was cut off at $19.5a_0$ beyond which the scattering solutions $R_{kl}^{\sigma}(r)$ were matched to the asymptotic form in Eq. (10). Our results were insensitive to the choice of this cut off radius as well as to the computational mesh. If the scattering phase shift per spin at zero energy $\delta_{l=0}^{\sigma}(\epsilon_k=0)=\pi$, bound states would appear in the s partial wave. A provision was made to look for the binding energy and wave function of this bound state.

III. SPIN- AND CHARGE-DENSITY DISTRIBUTIONS

Following the procedure outlined in Sec. II, we have computed self-consistent charge and spin distribution at metallic densities ($1\leq r_s\leq 5$) for different background polarizations ζ_0. In Figs. 1 and 2 we have plotted typical results for the normalized charge- and spin-density distributions, corresponding to $r_s=2$ and 5 and $\zeta_0=0.17$. We note a strong buildup of charge at the impurity, just as in the paramagnetic case[9] ($\zeta_0=0$).

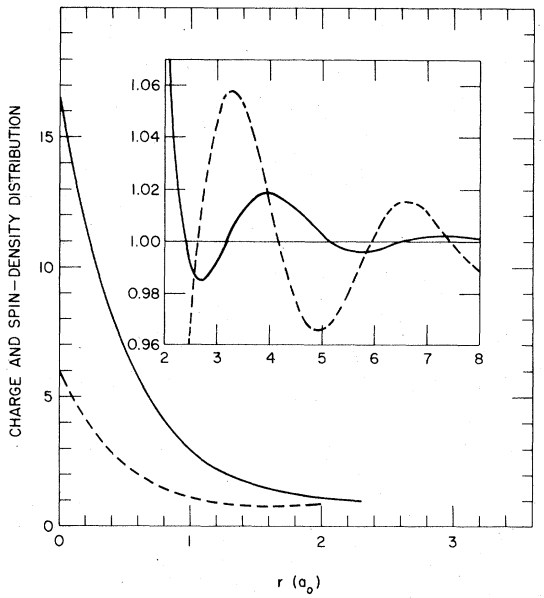

FIG. 1. Charge- and spin-density distribution around a positive muon in a spin-polarized electron gas with $r_s=2$ and $\zeta_0=0.17$. The solid and dashed curves correspond respectively to normalized charge density $n(r)/n_0$ and normalized spin density $n(r)\zeta(r)/n_0\zeta_0$.

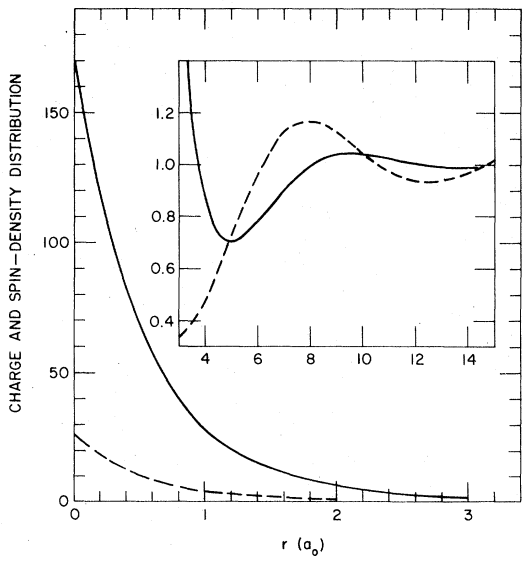

FIG. 2. Charge- and spin-density distribution around a positive muon in a spin-polarized electron gas with $r_s=5$ and $\zeta_0=0.17$. The rest of the legend is the same as that for Fig. 1.

As the bulk ζ_0 is increased from zero, the charge density at the origin $n(0)$ changes little from its paramagnetic value (less than 4% up to background polarization $\zeta_0 = 0.6$) with a drop of about (10–15)% in the purely ferromagnetic case ($\zeta_0 = 1$). Thus the contact charge density is largely fixed by the Coulomb singularity in the external potential, and the main effect of increasing bulk polarization on the density profiles is to make them more spread out in accord with the increased screening length in a polarized electron gas.[7]

Except for very large densities ($r_s \lesssim 1.9$) we have found for small polarizations ζ_0, the existence of bound states for both spins which are well extended in space. For a fixed ζ_0 the binding energy for each spin increases steadily as density decreases. For a fixed density the binding energy of the minority spin bound state increases with increasing ζ_0 while that of the majority spin decreases, and may eventually vanish at a critical ζ_0. In the purely ferromagnetic case ($\zeta_0 = 1$), of course no minority-spin states exist. The physical picture that emerges for the case of two bound states is that of an extended μ^- ion with an equally extended compensating hole in the background. The physical significance of the bound states, just as in the case of a proton in paramagnetic jellium, is uncertain due to lifetime effects and has been discussed earlier.[9]

Since it is the total density that has any physical meaning, the charge and spin densities in Figs. 1 and 2 contain contributions from both scattering and bound states. The total effective potentials for both spin-↑ and spin-↓ electrons at small r are dominated by the electrostatic term and fall off very rapidly. The binding energy for spin-↑ electron is consistently smaller than that of the spin-↓ electron. Consequently, the density of spin-↑ electron at the origin from bound-state contribution only is less than the corresponding value for spin-↓ electron. However, it is because of the influence of the scattering contribution that the total electron density of spin-↑ electrons at the origin, $n^+(0)$ is greater than $n^-(0)$. This implies that a positive muon simply enhances the ambient polarization at the origin without flipping its sign.

At distances far from the muon both charge and spin densities exhibit the usual Friedel oscillations. Note that not only the spin density at the origin is enhanced over the ambient polarization to a much lesser degree than the charge density, the Friedel oscillations lag by a phase of about $\frac{1}{2}\pi$. The amplitudes of the charge and spin oscillations for $r_s = 5$ are, however, significantly larger than those for $r_s = 2$. This results from a larger enhancement of spin and charge density at the μ^+ site for $r_s = 5$.

The dependence of the local spin polarization $\zeta(0)$ on the bulk ζ_0 is presented in Fig. 3. The reduction in $\zeta(0)$ is larger for small densities (large r_s), where a linear dependence is found (up to a point where the bulk gas becomes unstable and goes ferromagnetic). The reduction of the polarization is due to the tendency of the strongly attractive impurity to compensate for the background polarization.

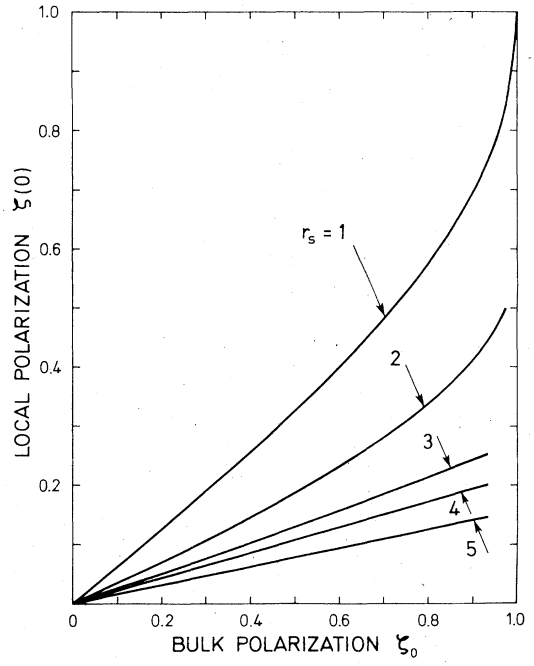

FIG. 3. Local spin polarization $\zeta(0)$ at a positive muon as a function of the bulk polarization ζ_0 of the host electron gas for different bulk densities r_s.

In Fig. 4 we have presented the electron charge- and spin-density enhancements at the origin over the unperturbed results for $1 \leq r_s \leq 5$. Since the charge density at the origin $n(0)$ depends somewhat on ζ_0, the spin-density enhancement $n(0)\zeta(0)/n_0\zeta_0$ can be represented by a universal curve in Fig. 4, which is valid to a 4% accuracy for $\zeta_0 < 0.6$ over the indicated density range. Note that while both the charge and spin-density enhancements increase steadily with r_s, the ratio of the spin-to-charge density enhancements decreases as the electron density decreases.

The charge- and spin-density enhancements of Fig. 4 can be well represented by the following cubic interpolation formulas:

$$n(0)/n_0 = 1.80 + 1.717 r_s + 0.370 r_s^2 + 1.193 r_s^3 \,, \tag{12}$$

$$n(0)\zeta(0)/n_0\zeta_0 = 5.267 - 5.060 r_s + 3.243 r_s^2 - 0.280 r_s^3 \tag{13}$$

for ($1 \leq r_s \leq 5$). It should be pointed out that the self-consistent spin-density enhancements at the μ^+ site in Eq. (13) is in good agreement with the predic-

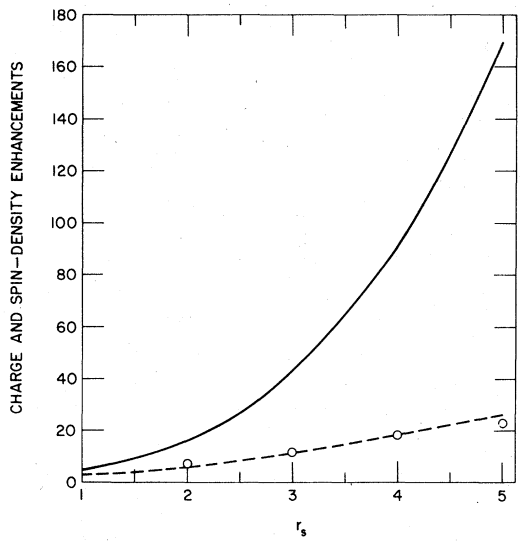

FIG. 4. Charge- and spin-density enhancements at a μ^+ site for bulk densities $1 \leq r_s \leq 5$. The solid and dashed curves correspond respectively to normalized charge density $n(0)/n_0$ and normalized spin density $n(0)\zeta(0)/n_0\zeta_0$. The open circles correspond to $V|\psi_{k_F}(0)|^2$ calculated in Ref. 9 for paramagnetic electron gas.

tions of a simple-model calculation by Meir[10] throughout the metallic density range. In his model,[10] Meir approximated the effective electron-muon potential by the Hulthen form $-e^2\lambda/(e^{\lambda r}-1)$, where the screening parameter λ is determined from the Friedel sum rule. This agreement implies that the enhancement in the spin density at the muon site is primarily due to the singularity at the origin in the effective electron-muon potential.

To analyze the spin-density enhancements further, we write the spin density as[11]

$$n^+(0)-n^-(0)=\sum_k |\psi_k(0)|^2 \times [f(\epsilon_k+\mu_B H_0)-f(\epsilon_k-\mu_B H_0)] \ , \tag{14}$$

where the symbols have their usual meaning. From Eq. (14) it follows that

$$\frac{n^+(0)-n^-(0)}{n_{+0}-n_{-0}} \cong |\psi_{k_F}(0)|^2 V \ , \tag{15}$$

V being the volume of the system. Thus for small initial polarization, the spin-density enhancement computed by considering an impurity in an initially spin-polarized background is the same as the wave function density for $k=k_F$ evaluated in a corresponding paramagnetic calculation. To see how these two quantities compare, we have also plotted in Fig. 4 our earlier result[9] for $V|\psi_{k_F}(0)|^2$. The small discrepancy between these two results could be partly due to the use of a different form of exchange-correlation[7] potential in the present calculation.

Thus, the conventional expression for the Knight shift K,

$$K=\tfrac{8}{3}\pi\chi|\psi_{k_F}(0)|^2 V \tag{16}$$

can also be rewritten

$$K=\tfrac{8}{3}\pi\chi[n^+(0)-n^-(0)]/(n_{+0}-n_{-0}) \ , \tag{17}$$

where χ is the spin susceptibility of the system. It should again be emphasized that in the HKS formalism only the electron density can be assigned physical meaning. However, the near equality of $V|\psi_{k_F}(0)|^2$ and $[n^+(0)-n^-(0)]/(n_{+0}-n_{-0})$ suggests that the use of either expression (16) or (17) to compute Knight shift is acceptable.

IV. HYPERFINE FIELDS

The foregoing considerations are strictly valid for a spin-polarized jellium metal. Given the knowledge of ambient polarizations we can, as we have seen, calculate the hyperfine field. Do these considerations have any semblance of reality when applied to ferromagnetic metals like Fe, Co, Ni, and Gd? The obvious answer is: It is perhaps unlikely. Nonetheless, we shall assume them to be valid and examine the consequences.

We shall first consider the case of the more common $3d$ transition-metal ferromagnets. We assume a localized picture for ferromagnetism in which the d electrons are localized and carry almost all of the measured magnetic moment. The s electrons are free and carry a very small polarization due to s-d exchange. The interstitial spin polarization is, therefore, in this model entirely due to s electrons. In this picture the number of s electrons per magnetic ion is 1 for Fe and Co and 0.6 for Ni. This corresponds to r_s values of 2.7, 2.6, and 3.1 for Fe, Co, and Ni, respectively.

Using the ambient polarization density as determined from neutron scattering data[12–15] in Table I, and the calculated spin-density enhancement from Eq. (13), the results for the hyperfine fields are given in Table I. It is seen that the calculated fields are an order of magnitude larger than those given by experiment.[16–19]

This large discrepancy between theory and experiment raises serious doubts regarding the validity of the model used for calculating the hyperfine fields. It appears essential to take into account the itinerant character of the d electrons. This would mean that

TABLE I. Hyperfine fields at an interstitial μ^+ site in ferromagnetic Fe, Co, Ni, and Gd.

Host metal	r_s	Ambient spin density $n_0\zeta_0$ ($\mu_B/\text{Å}^3$)	B_{loc}(kOe)[a]	$B_{\text{hf}}^{\text{expt}}$ (kOe)	$B_{\text{hf}}^{\text{theo}}$ (kOe)[b]
Fe	2.7	+0.106/−0.215 (Ref. 12)	+8.4/−16.8 (Ref. 16)	−11.1	+82/−164
Co	2.6	−0.12 (Ref. 13)	−9.3 (Ref. 17)	−6.2	−84
Ni	3.1	−0.0085 (Ref. 14)	−0.66 (Ref. 18)	−0.64	−8.2
Gd	2.6	−0.037 (Ref. 15)	−2.9 (Ref. 19)	−7.5	−26

[a] $B_{\text{loc}} = -\frac{8}{3}\pi\mu_B n_0\zeta_0$ (assuming the positive muon does not perturb the ambient polarization).

[b] $B_{\text{hf}}^{\text{theo}} = -\frac{8}{3}\pi\mu_B n(0)\zeta(0)/n_0\zeta_0$ (assuming the ambient spin density in column 3 as given by neutron scattering experiments to be the true average spin polarization a muon "sees"). In Fe, there is no unique value available for the ambient density (see Ref. 12). Thus, the two theoretical values of B_{hf} at μ^+ site in Fe is due to the two different choices of the ambient spin density.

one has to set up a HKS scheme in which there are two species of electrons (the "light" s electron and the "heavy" d electron) and take into account the exchange coupling between them. Even with this simple modification, a fully self-consistent calculation would require considerable numerical effort.

If one assumes that the s electrons are little or not polarized and the interstitial moment density is due to the quasilocalized d electrons which remain unaffected by the presence of a muon, then the ambient spin density gives rise to a hyperfine field given in column 4 of Table I. Note that the agreement between these values and experiment is better than a factor of 2. Such an argument has been put forth by Petzinger and Munjal[4] for the case of Ni. This argument may appear plausible in view of the fact that the s electrons being "light" can readily respond to the presence of the muon. However, it should be kept in mind that the number of quasilocalized d electrons is an order of magnitude larger than the number of s electrons. It is, therefore, unclear why the s electrons would dominate the screening process. The above agreement could thus be fortuitous.

We shall now consider a somewhat simpler case of ferromagnetic Gd. It is a rare-earth metal in which the unpaired $4f$ orbitals are thought to be highly localized and carry a magnetic moment of $7\mu_B$. The saturation magnetic moment[15] per atom is $7.63\mu_B$. Therefore, the remaining $0.63\mu_B$ must be attributed to the three sp electrons in the conduction band. Both band-structure theory[20] and experiment[15] agree that the polarization density at the c site is *negative* in sign, although they differ in magnitude by about a factor of 20 ($-0.037\mu_B/\text{Å}^3$ from experiment[15] and $-0.002\mu_B/\text{Å}^3$ from band structure[20]). Using the experimental value[15] for the ambient polarization and $r_s = 2.6$, we obtain a hyperfine field (given in Table I) which is a factor of four greater than the experimental value.[19] On the other hand, if one were to use the band structure value,[20] the hyperfine field will be smaller than the experimental value[19] by a factor of 5. In view of the fact that the model described here ought to be applicable to metals like Gd, one would like to understand the source of this discrepancy. Two such obvious sources come to mind: (i) the accuracy of the neutron form factor data and (ii) the neglect of the zero-point vibration for the muon. The quoted error in the experimental results of the ambient spin density is ±50% The various sources of error in the neutron experiment have been discussed by earlier authors.[15,20] Unless the experimental data are grossly in error, it is hard to understand the discrepancy in the hyperfine field. Because of the large amplitude ($\sim 1a_0$) of the zero-point vibration, the muon could make excursion into the region where the ambient spin density is positive. Since the total magnetic moment carried by the conduction electrons is $+0.63\mu_B$ per atom, it is certain that there are large regions of space where the spin density is positive. We have not considered the effect of this inhomogeneity on the spin enhancement at the muon site.

This paper should be regarded as a preliminary attempt towards an *a priori* understanding of a point charge impurity in ferromagnetic systems. Obviously this understanding is linked to an understanding of ferromagnetism. A more ambitious theoretical program would be one in which one considers the positive muon and a shell of surrounding magnetic ions forming a super unit cell. With this super lattice structure, energy bands and wave functions have to be obtained self-consistently. The effect of exchange and correlation can be incorporated through the spin-density functional formalism. Easy as it may sound, the problem poses tremendous numerical complications. Unfortunately, this has to be done if one wants to achieve meaningful quantitative comparison between theoretical and experimental hyperfine fields at interstitial μ^+ sites in transition-metal ferromagnets.

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