# Magnetic form factor for itinerant-electron systems

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First-principles calculations of the itinerant-electron magnetic form factor for ferromagnets which include both the spin and orbital contributions have been completed for ferromagnetic nickel and iron. To our knowledge, these are the first calculations of this type for 3*d* ferromagnets. These calculations also include, by necessity, the spin-orbit interaction. Numerical results based on spin polarized bands used previously by Cooke *et al.* [Phys. Rev. B **21**, 4118 (1980)] have been found to be in good agreement with experiment while results based on local density bands were found to be in good agreement for iron but not for nickel. In addition, it has been shown that if the energy dependence of the radial part of the wave functions can be ignored then the dominant terms in the itinerant-electron form factor can be written in a form almost identical to the atomic form factor. This provides an explanation for why fits to the atomic form factor have been so successful.

## INTRODUCTION

The magnetic form factor measured by elastic neutron-scattering techniques yields direct information about the Fourier transform of the ground-state spin density and, therefore, about the electronic wave functions of a solid. There are three distinct contributions to the form factor. The largest component is essentially the Fourier transform of the electronic spin density. The second arises from the orbital motion of the electrons. The third contribution results from the core electrons which occupy closed shells and are virtually unaffected by local environmental effects. Because many techniques, such as x-ray photoemission, do not probe solely the ground state, form-factor measurements are of particular interest for local spin-density theory. In the past, experimentally determined form factors for magnetic solids have been analyzed at least partially in terms of a theoretical expression derived for atomiclike systems.[1] The atomic theory is adapted to itinerant-electron magnets somewhat arbitrarily by dividing the electronic system into "nonlocalized" and "localized" parts. The nonlocalized electrons are assumed to be uniformly distributed throughout the crystal, while the localized electrons combine to form an appropriate atomic configuration. The total form factor, normalized to unity at $K=0$, obtained from this model can be written in the general form

$$\frac{1}{\hbar}F(\mathbf{K})=M_{NL}F_{NL}(\mathbf{K})+M_L F_L(\mathbf{K})+M_{\rm orb}F_{\rm orb}(\mathbf{K}) +F_{\rm core}(\mathbf{K}) , \quad (1)$$

where $M$ is the fraction of the total moment for each case, $F(\mathbf{K})$ is the corresponding form factor, and $\mathbf{K}$ is restricted to reciprocal-lattice vectors. The numerically significant contributions to the form factors used in this analysis are given by

$$F_{NL}(\mathbf{K})=\delta_{\mathbf{K},0} , \quad (2)$$

$$F_L(\mathbf{K})=\langle j_0(\mathbf{K})\rangle_d+(\tfrac{5}{2}\gamma-1)\langle j_4(\mathbf{K})\rangle_d A(\mathbf{K}) , \quad (3)$$

$$F_{\rm orb}(\mathbf{K})=\langle g_0(\mathbf{K})\rangle_d-\tfrac{1}{2}\langle g_2(\mathbf{K})\rangle , \quad (4)$$

where $\gamma$ is the Weiss-Freeman parameter (the fraction of the 3*d* electrons in $e_g$ orbitals) and

$$\langle j_l(K)\rangle_d=\int j_l(Kr)R_d^2(r)r^2dr , \quad (5)$$

$$\langle g_l(K)\rangle_d=\int g_l(Kr)R_d^2(r)r^2dr . \quad (6)$$

$R_d(r)$ is the atomic radial function for *d* symmetry. The $j_l(x)$ are spherical Bessel functions and the $g_l(x)$ are similar functions originally defined by Trammell.[2] The asymmetry factor is

$$A(\hat{\mathbf{K}})=\tfrac{5}{2}[\hat{\mathbf{K}}_x^4+\hat{\mathbf{K}}_y^4+\hat{\mathbf{K}}_z^4-\tfrac{3}{5}] , \quad (7)$$

where $\hat{\mathbf{K}}$ indicates a unit vector. As shown by Watson and Freeman,[3] the core contribution results from relatively small differences between the radial functions for opposite-spin core electrons; there is, of course, no net spin associated with the filled core. The magnitude of this is term is comparable with experimental uncertainty and will be neglected.

The first two terms in Eq. (1) comprise the spin contribution. The form of the first term arises because of the assumed uniform distribution of the "nonlocal" electrons. The local term was first derived by Weiss and Freeman[4] and represents the contribution from the 3*d* electrons. The orbital contribution was first derived by Blume[5] and was obtained by placing the 3*d* atomic configuration into a crystal field consistent with the symmetry of the solid. Blume obtained an additional asymmetry term (proportional to $A(K)$ in the orbital contribution [Eq. (4)]), but this is small and generally neglected.

The moment fractions are usually expressed in terms of the atomic *g* factor and the fraction of the *d*-like spin moment which is nonlocal, $\alpha$:

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$$M_{NL}=2\alpha/g \ , \tag{8}$$

$$M_L=2(1-\alpha)/g \ , \tag{9}$$

$$M_{\rm orb}=(g-2)/g \ . \tag{10}$$

Excellent fits to experimentally determined form factors have been obtained for both nickel and iron by varying the atomic configuration and the parameters $\alpha$ and $g$.[1] There are, however, a number of conceptual problems which arise when this theory is applied to itinerant-electron systems. These problems are all traceable to the concept of using an atomic configuration to describe the electronic properties of a metallic system. In addition, because of its atomistic nature, the expression in Eq. (1) cannot be used with any degree of certainty to test proposed spin-polarized band structures for itinerant-electron magnets.

A number of calculations of the spin part have been reported for nickel and iron.[6,7] The purpose of this paper is to investigate the total magnetic form factor appropriate for itinerant-electron magnets and to present some numerical results for nickel and iron. The paper is divided into six parts. The first outlines the general theory, the second and third give some results for the spin and orbit terms, respectively, the fourth provides a comparison with the atomistic theory, the fifth presents some numerical results, and the last gives a summary and conclusions.

## GENERAL THEORY

Simply stated, the derivation of the form factor for itinerant-electron systems can be reduced to the evaluation of

$$\frac{1}{\hbar}F(\mathbf{K})=F_{\rm spin}(\mathbf{K})+F_{\rm orb}(\mathbf{K}) \ , \tag{11}$$

$$F_{\rm spin}(\mathbf{K})=[\langle \mathbf{s}_{\mathbf{K}}\rangle-\hat{\mathbf{K}}(\langle \mathbf{s}_{\mathbf{k}}\rangle\cdot\hat{\mathbf{K}})]\cdot\hat{\mathbf{m}} \ , \tag{12}$$

$$\langle \mathbf{s}_{\mathbf{K}}\rangle=\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\langle \mathbf{s}_i\rangle \ , \tag{13}$$

$$F_{\rm orb}(\mathbf{K})=-\frac{i(\hat{\mathbf{K}}\times\langle \mathbf{p}_{\mathbf{K}}\rangle)\cdot\hat{\mathbf{m}}}{\hbar|\mathbf{K}|} \ , \tag{14}$$

$$\langle \mathbf{p}_{\mathbf{K}}\rangle=\sum_j e^{i\mathbf{K}\cdot\mathbf{r}_j}\mathbf{p}_j \ , \tag{15}$$

where $s_j$ and $p_j$ are the spin and linear momentum operators, respectively, of the $j$th electron and $\hat{\mathbf{m}}$ is a unit vector along the moment direction ($\mathbf{m}=\langle \mathbf{s}_{\mathbf{K}=0}\rangle$). This is the quantity that is obtained from elastic neutron-scattering experiments using polarization analysis. Usually, the moment direction is chosen to be perpendicular to the scattering plane, in which case the second term in Eq. (12) is zero. This convention will be adopted throughout this paper. The more general case can be treated using similar techniques.

It is straightforward to show for cubic systems that the so-called orbital term in Eq. (11) is zero unless the spin-orbit interaction is taken into account. Since this interaction is known to be non-negligible in many itinerant-electron magnets, it must be included in the calculation. Inclusion of the spin-orbit interaction leads to complex wave functions, a mixing of opposite-spin states, and a reduction in symmetry.

The expectation values in Eq. (11) are evaluated in terms of the single-particle band structure wave function

$$\psi_{n\mathbf{k}}(\mathbf{r})=\sum_{\mu\sigma} a_{n\mu\sigma}(\mathbf{k})\phi_\mu^\sigma(\mathbf{r},E_{n\mathbf{k}})\chi_\sigma \ , \tag{16}$$

where $\mu$ is a symmetry index ($\mu\equiv l_\mu,m_\mu$) which runs over $s$, $p$, and $d$ terms and $\sigma$ is the spin index. The $\{\phi_\mu^\sigma(\mathbf{r},E)\}$ are energy-dependent symmetry orbitals, $\chi_\sigma$ is a spinor, and the $\{a_{n\mu\sigma}(\mathbf{k})\}$ are expansion coefficients. For suitable choices of $\phi_\mu^\sigma(\mathbf{r},E)$, this general form of the wave function is valid for interpolation schemes such as those proposed by Slater and Koster[8] (SK) or Hodges, Ehrenreich, and Lang (HEL).[9] It is also appropriate, within the muffin-tin sphere, for wave functions obtained from KKR band-structure calculations.

Then, for any operator $O$,

$$\langle O(\mathbf{K})\rangle=\sum_{\substack{\mu\nu\\ \sigma}}\int \rho_{\mu\nu}^\sigma(E)f(E)D_{\mu\nu}^\sigma(\mathbf{K},E)dE \ , \tag{17}$$

where

$$\rho_{\mu\nu}^\sigma(E)=\frac{1}{N}\sum_{n\mathbf{k}}\bar{a}_{n\mu\sigma}(\mathbf{k})a_{m\nu\sigma}(\mathbf{k})\delta(E-E_{n\mathbf{k}}) \ , \tag{18}$$

$$D_{\mu\nu}^\sigma(\mathbf{K},E)=\int \bar{\phi}_\mu^\sigma(\mathbf{r},E)\bar{\chi}_\sigma O\chi_\sigma\phi_\nu^\sigma(\mathbf{r},E)d^3r \ , \tag{19}$$

and $f(E)$ is the Fermi occupation factor.

If the symmetry orbitals $\phi_\mu^\sigma$ can be written in the general form

$$\phi_\mu^\sigma(\mathbf{r},E)=(i)^{l_\mu}R_\mu(r,E)Y_\mu(\hat{\mathbf{r}}) \ , \tag{20}$$

where $R_\mu(r,E)$ is an energy-dependent radial function and $Y_\mu(\hat{r})$ is a spherical harmonic, and the identity

$$e^{iK\cdot r}=4\pi\sum_\mu (i)^{l_\mu}j_{l_\mu}(Kr)Y_\mu(\hat{\mathbf{r}})Y_\mu(\hat{\mathbf{K}}) \tag{21}$$

is used, then $D_{\mu\nu}^\sigma$, in Eq. (15), can be evaluated in a straightforward way for both the spin and orbital cases. The $Y_\mu$ are chosen to be real spherical harmonics, which makes the expansion coefficients in Eq. (16) complex. The form for $\phi_\mu^\sigma$ given in Eq. (20) will be used throughout this paper.

Inclusion of the spin-orbit interaction affects the form factor in two ways. First, the explicit form of the form factor depends on the direction of the magnetic-moment direction relative to the crystalline axes which is defined by the conventional Euler angles $\theta$ and $\phi$. If, for example, the moment direction is restricted to the crystalline $z$ axis ($\theta=0$), the spin-orbit interaction reduces the crystal symmetry for cubic ferromagnets from $O_h$ to $C_{4h}$; only inversion symmetry remains for the general case. Second, additional terms appear in the expression which are generated by the spin-orbit interaction; these terms depend explicitly on the complex nature of the electronic wave functions. This can best be demonstrated by referring to some explicit results.

## SPIN FORM FACTOR

The spin part of the form factor is calculated from Eqs. (12) and (17) with $\langle O \rangle = \langle \mathbf{s}_{\mathbf{K}} \rangle$ and

$$D^{\sigma}_{\mu\nu}(\mathbf{K},E)=\frac{m_\sigma}{2}\int \phi^{\sigma}_{\mu}(\mathbf{r},E)e^{i\mathbf{K}\cdot\mathbf{r}}\phi^{\sigma}_{\nu}(\mathbf{r},E)d^3r \ , \tag{22}$$

with

$$m_\sigma=\pm 1 \quad \text{for } \sigma=\uparrow,\downarrow \ . \tag{23}$$

By using the identity given in Eq. (21) and the form for $\phi^{\sigma}_{\mu}$ given in Eq. (20), it follows that $D^{\sigma}_{\mu\nu}(\mathbf{K},E)$ can be written in the form

$$D^{\sigma}_{\mu\nu}(\mathbf{K},E)=2\pi m_\sigma \sum_{\eta}\langle j_{l_\eta}(K,E)\rangle^{\sigma}_{l_\mu l_\nu} C^{\eta}_{\mu,\nu} Y_\eta(\hat{\mathbf{K}}) \ , \tag{24}$$

where

$$\langle j_{l_\eta}(K,E)\rangle^{\sigma}_{l_\mu l_\nu}=\int r^2 R^{\sigma}_{l_\mu}(r,E) j_{l_\eta}(Kr) R^{\sigma}_{l_\nu}(r,E)dr \tag{25}$$

and

$$C^{\eta}_{\mu\nu}=(i)^{-l_\mu+l_\nu+l_\eta}\int Y_\mu(\hat{\mathbf{r}})Y_\eta(\hat{\mathbf{r}})Y_\nu(\hat{\mathbf{r}})d\hat{\mathbf{r}} \ . \tag{26}$$

The spin form factor can be numerically evaluated by calculating each of the individual terms and summing over the appropriate indices. Alternately, substitution of Eq. (22) into Eqs. (17) and (12) and simplifying gives

$$F_{\text{spin}}(\mathbf{K})=[1-(\hat{\mathbf{m}}\cdot\hat{\mathbf{K}})^2]\sum_{\sigma}\frac{m_\sigma}{2}[I^{\sigma}_1(K,E)+I_2(K,E)Y_{4,0}(\hat{\mathbf{K}})+I^{\sigma}_3(K,E)Y_{4,4}(\hat{\mathbf{K}})+\cdots]f(E)dE \ , \tag{27}$$

where

$$\hat{\mathbf{m}}\cdot\hat{\mathbf{K}}=\sin\theta\cos\phi\,\hat{\mathbf{K}}_x+\sin\theta\sin\phi\,\hat{\mathbf{K}}_y+\cos\theta\,\hat{\mathbf{K}}_z \ , \tag{28}$$

$$I^{\sigma}_1(K,E)=\sum_{\mu=1}^{9}\langle j_0(K,E)\rangle^{\sigma}_{l_\mu,l_\mu}\rho^{\sigma}_{\mu,\mu}(E) \ , \tag{29}$$

$$I^{\sigma}_2(K,E)=\frac{2\sqrt{\pi}}{7}(\rho^{\sigma}_{5,5}-4\rho^{\sigma}_{6,6}-4\rho^{\sigma}_{7,7}+\rho^{\sigma}_{8,8}+6\rho^{\sigma}_{9,9}) \ , \tag{30}$$

with

$$I^{\sigma}_3(K,E)=2\sqrt{5\pi/7}(\rho^{\sigma}_{8,8}-\rho^{\sigma}_{5,5}) \ . \tag{31}$$

The connection between the symmetry indices $\mu$ in Eqs. (28)–(31) and the symmetry orbitals is given in Table I. The $Y_{lm}$ are real spherical harmonics:

$$Y_{4,0}(\hat{\mathbf{K}})=\frac{3}{8}\frac{1}{\sqrt{4\pi}}(35\hat{\mathbf{K}}_z^4-30\hat{\mathbf{K}}_z^2+3) \ , \tag{32}$$

$$Y_{4,4}(\hat{\mathbf{K}})=\left[\frac{315}{256\pi}\right]^{1/2}(\hat{\mathbf{K}}_x^4-6\hat{\mathbf{K}}_x^2\hat{\mathbf{K}}_y^2+\hat{\mathbf{K}}_y^4) \ . \tag{33}$$

The series in Eq. (27) terminates after the third term if the spin-orbit interaction is neglected. For this case it follows from symmetry arguments that $\rho^{\sigma}_{\mu\nu}(E)=\delta_{\mu,\nu}\rho^{\sigma}_{\mu\mu}(E)$ and only the terms given explicitly in Eq. (27) are nonzero. If plane waves are used to describe the non-$d$ symmetry terms, then the non-$d$ part of the form factor reduces to the result given by HEL.[9]

TABLE I. Convention relating symmetry index $\mu$ to the symmetry orbital $\phi_\mu(\mathbf{r},E)=R_\mu(r,E)Y_\mu(\mathbf{r})$.

| $\mu$ | $Y_\mu(\mathbf{r})$ |
|---|---|
| 1 | constant |
| 2 | $x/r$ |
| 3 | $y/r$ |
| 4 | $z/r$ |
| 5 | $xy/r^2$ |
| 6 | $yz/r^2$ |
| 7 | $xz/r^2$ |
| 8 | $(x^2-y^2)/r^2$ |
| 9 | $(3z^2-r^2)/r^2$ |

## ORBITAL FORM FACTOR

Like the spin form factor, the orbital form factor can be calculated from Eqs. (14) and (17), but with $\langle O \rangle = \langle \mathbf{p}_{\mathbf{K}} \rangle$ and

$$\mathbf{D}^{\sigma}_{\mu\nu}(\mathbf{K},E)=-i\int \phi^{\sigma}_{\mu}(r)e^{i\mathbf{K}\cdot\mathbf{r}}\nabla\phi^{\sigma}_{\nu}(r)d^3r \ . \tag{34}$$

The orbital form factor can be written in a more convenient form by making use of symmetry arguments, the fact that the $\mathbf{D}^{\sigma}_{\mu\nu}$ are real, and that only the $s$-$d$ and $d$-$d$ terms are nonzero. It follows that

$$\hat{\mathbf{K}}\times[\mathbf{D}^{\sigma}_{\mu\nu}(\mathbf{K},E)+\mathbf{D}^{\sigma}_{\nu\mu}(\mathbf{K},E)]=0 \tag{35}$$

and, therefore,

$$F_{\text{orb}}(\mathbf{K})=\sum_{\substack{\mu\nu\\ \sigma}}\int\frac{\hat{\mathbf{m}}\cdot[\hat{\mathbf{K}}\times\mathbf{D}^{\sigma}_{\mu\nu}(\mathbf{K},E)]}{|\mathbf{K}|}f(E)\tilde{\rho}^{\sigma}_{\mu\nu}(E)dE \ , \tag{36}$$

where $\tilde{\rho}^{\sigma}_{\mu\nu}(E)$ is the imaginary part of $\rho^{\sigma}_{\mu\nu}(E)$. If the spin-orbit interaction is neglected, all quantities are real and the orbital contribution clearly vanishes.

As in the spin case, an expression for $\mathbf{D}^{\sigma}_{\mu\nu}$ suitable for numerical evaluation can be derived. Even though only $s$-$d$ and $d$-$d$ symmetry terms are nonzero, this expression is inherently more complex than the spin case. The $d$-$d$ contributions, which are significantly greater than the $s$-$d$ contributions, can be written in the form

$$F_{\text{orb}}(\mathbf{K})=\hat{\mathbf{m}}\cdot\hat{\mathbf{K}}\times\sum_{\eta,\sigma}\mathbf{S}^{\sigma}_{\eta}(\mathbf{K},E)\langle J_{l_\eta}(K,E)\rangle^{\sigma}_{d}f(E)dE \ . \tag{37}$$

The various quantities are defined in the Appendix. Only terms of order $l_\eta=1$ and 3 contribute. Substitution of Eq. (A13) into Eq. (37) and simplifying gives, for the $l_\eta=1$ terms,

$$F_{\rm orb}(\mathbf{K})=B_1(K)\{\sin\theta[\sin\phi(1-\hat{\mathbf{K}}_y^2)-\cos\phi\,\hat{\mathbf{K}}_x\hat{\mathbf{K}}_y]-\cos\theta\,\hat{\mathbf{K}}_y\hat{\mathbf{K}}_z\}$$
$$+B_2(K)\{\sin\theta[\cos\phi(1-\hat{\mathbf{K}}_x^2)-\sin\phi\,\hat{\mathbf{K}}_y\hat{\mathbf{K}}_x]-\cos\theta\,\hat{\mathbf{K}}_x\hat{\mathbf{K}}_z\}$$
$$+B_3(K)[\cos\theta(1-\hat{\mathbf{K}}_z^2)-\sin\theta(\cos\phi\,\hat{\mathbf{K}}_x\hat{\mathbf{K}}_y+\sin\phi\,\hat{\mathbf{K}}_y\hat{\mathbf{K}}_z)]\ , \tag{38}$$

where

$$B_i(G)=\sum_\sigma\int\langle J_1(K,E)\rangle_d^\sigma f(E)A_i^\sigma(E)dE \qquad (i=1,3)\ , \tag{39}$$

$$\langle J_1\rangle_d^\sigma=\langle j_0\rangle_{2,2}^\sigma+\langle j_2\rangle_{2,2}^\sigma\ , \tag{40}$$

$$A_1^\sigma(E)=\tilde{\rho}_{7,8}^\sigma+\tilde{\rho}_{6,5}^\sigma+\sqrt{3}\tilde{\rho}_{9,7}^\sigma\ , \tag{41}$$

$$A_2^\sigma(E)=\tilde{\rho}_{6,8}^\sigma+\tilde{\rho}_{5,7}^\sigma+\sqrt{3}\tilde{\rho}_{6,9}^\sigma\ , \tag{42}$$

$$A_3^\sigma=\tilde{\rho}_{7,6}^\sigma+2\tilde{\rho}_{8,5}^\sigma\ . \tag{43}$$

A similar result can be derived for the $l_\eta=3$ order terms, few enough to work out by hand, but too many to reproduce here.

## CONNECTION WITH THE ATOMIC MODEL

Calculations based on the atomic model outlined above for conventional experimental geometry ($\theta\to0$, $\hat{\mathbf{K}}_z\to0$) require as input various adjustable parameters, e.g., $\alpha$, $\gamma$, and $g$, and the electronic wave function determined from a given atomic configuration. The best fit to form-factor data is obtained by varying the parameters and the atomic configuration. The energy-dependent wave function is usually evaluated at the Fermi energy.

Comparison of the itinerant spin and orbital results given in Eqs. (27) and (38) with the atomic model results given in Eqs. (1)–(7) reveals a number of similarities. In fact, the expressions can be made identical in form provided three approximations are made for the itinerant case. First, as in the atomic model, the energy dependence of the wave functions is ignored, e.g., evaluated at the Fermi energy. Second, the spin-orbit interaction is neglected when calculating the spin part of the form factor. The system then has cubic symmetry, and it follows that

$$\rho_{5,5}^\sigma=\rho_{6,6}^\sigma=\rho_{7,7}^\sigma=\tfrac{1}{3}n_{t_{2g}}^\sigma$$

and

$$\rho_{8,8}^\sigma=\rho_{9,9}^\sigma=\tfrac{1}{2}n_{e_g}^\sigma\ ,$$

where $n_{t_{2g}}^\sigma$ and $n_{e_g}^\sigma$ are the number of electrons with spin $\sigma$ and $t_{2g}$ and $e_g$ symmetry, respectively. The third approximation is to neglect the non-$d$ symmetry terms, which are very small. Note that $\alpha=0$ here since the full moment is included in the calculation. This leads to identical forms for the spin part of the form factor. The equivalence of the orbital contributions follows from the identity

$$\langle g_0\rangle_d-\tfrac{1}{2}\langle g_2\rangle=\langle j_0\rangle_d+\langle j_2\rangle_d\ .$$

Comparison of the itinerant and atomic expressions also yields explicit results for the various parameters in terms of the electronic band structure. These are

$$m_{\rm spin}=\sum_\sigma\frac{m_\sigma}{2}(n_{t_{2g}}^\sigma+n_{e_g}^\sigma)\ ,$$

$$m_{\rm orb}=\sum_\sigma(\tilde{\rho}_{3,2}^\sigma+2\tilde{\rho}_{4,1}^\sigma)\ ,$$

$$g=2\left[1+\frac{m_{\rm orb}}{m_{\rm spin}}\right]\ ,$$

$$\gamma=\frac{\sum_\sigma(m_\sigma/2)n_{e_g}^\sigma}{m_{\rm spin}}\ .$$

The $\tilde{\rho}_{\mu\nu}^\sigma$ and $\langle j_l\rangle_d$ are calculated from the electronic energies and wave functions obtained from the band structure. It could be argued that the fitting procedure used in the atomic model works because the general structure of the form factor is correct; i.e., the fitting procedure attempts to mimic the results that would have been obtained from the itinerant model. This argument, of course, relies on the approximations used above. It also assumes that the itinerant model can reproduce the experimental form factor. In order to determine if this is the case, the form factor must be numerically evaluated.

## NUMERICAL RESULTS

The calculation of the magnetic form factor for itinerant systems requires as input the full self-consistent spin-polarized band structure. Unfortunately, there are at present no published band structures for transition-metal magnets which include the spin-orbit interaction. In order to proceed, we have incorporated the spin-orbit interaction into published band structures by use of an interpolation approach suggested by HEL.[9] Although this procedure is not exact, it does provide reasonable predictions in the absence of first-principles results.

The expression for the form factor given in Eq. (11) was evaluated using the general expression given in Eq. (17) and results given in Eqs. (24)–(25) for the spin case and in Eqs. (A5), (A6), and (A10) for the orbital case. The Brillouin-zone sums were obtained using the tetrahedron method. The Wigner-Seitz sphere approximation to the unit cell was used to evaluate the radial integrals, and the energy dependence of the radial integrals was treated in two ways. First, the energy dependence of the radial integrals was fitted to a polynomial in energy. The second method was to evaluate the radial integrals at the Fermi energy, which amounts to neglecting the energy dependence altogether. Twelve bands were used in the calculation.

Two different types of spin-polarized band structures

were used. One type consisted of energy bands used previously in calculations of the spin dynamics of nickel and iron [Cooke, Lynn, and Davis (CLD)].[10] These bands yielded excellent agreement with results from inelastic neutron-scattering experiments as well as predictions of unusual spin-wave behavior, which was subsequently confirmed. The other type were spin-polarized bands published by Moruzzi, Janak, and Williams (MLW).[11] based on local density theory. The radial functions were determined from appropriate potentials in each case. Also, the bands were fitted to a HEL interpolation scheme for nickel and a Slater-Koster interpolation scheme for ions, and then the spin-orbit interaction was included using the method proposed by HEL.[9] The spin-orbit parameter used for nickel was suggested by HEL,[9] and a suitably scaled parameter was used for iron. These values are close to the point-charge predictions for these materials.

Numerical results which include the energy dependence of the radial functions for nickel are given in Figs. 1 and 2 for the CLD and MJW bands, respectively. Corresponding results for iron are given in Figs. 3 and 4. In these figures, $\theta$ is half the scattering angle and $\lambda$ is the neutron wavelength, and only data points corresponding to the case where the moment direction is parallel to the $z$ axis of the crystal are given. The experimental and theoretical form factors are normalized to unity at (000) and the energy dependence of the wave function is included. The results based on the band structures that produced good agreement with the spin dynamics of nickel and iron also yield good agreement with the form factor. Another interesting result obtained from these calculations is that the energy dependence of the wave functions is important. A comparison of the form factor for iron calculated with the energy-dependent wave functions and with the wave functions evalauted at the Fermi energy is shown in Fig. 5.

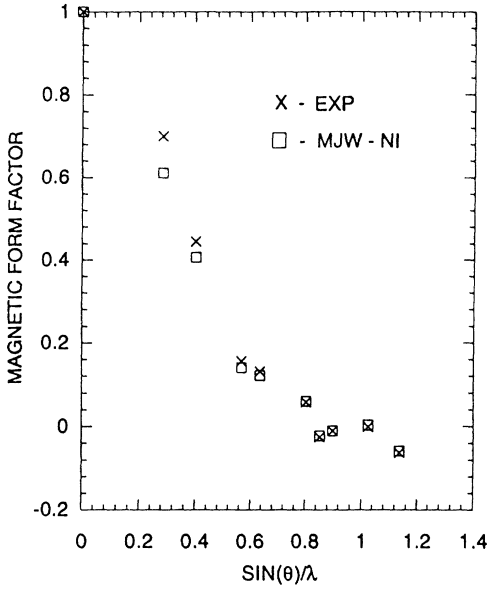


FIG. 2. Comparison of theoretical and experimental form factors normalized to unity at (000) for nickel ($\lambda$ in Å). Theory result based on MJW local density bands; experimental results from Mook (Ref. 12).

The bands based on local density theory yield good agreement with iron, but not for nickel, at least for the MJW bands. It is worth noting that the orbital contribution to the form factor is much less sensitive than the spin part to the band structure used (see Tables II and III for Ni; effects of a similar magnitude also occur in Fe). Furthermore, the orbital part contributes generally about

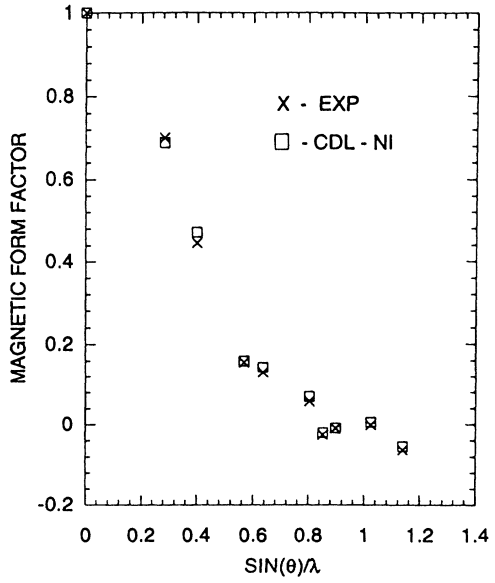


FIG. 1. Comparison of theoretical and experimental form factors normalized to unity at (000) for nickel ($\lambda$ in Å). Theory result based on CLD bands; experimental results from Mook (Ref. 12).

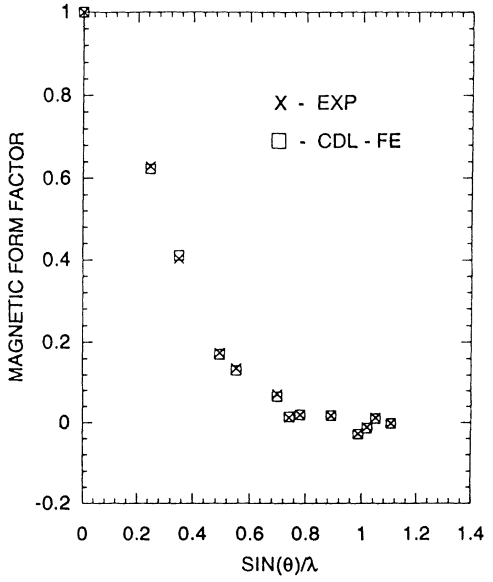


FIG. 3. Comparison of theoretical and experimental form factors normalized to unity at (000) for iron ($\lambda$ in Å). Theory result based on CLD bands; experimental results from Shull and Yamada (Ref. 13).

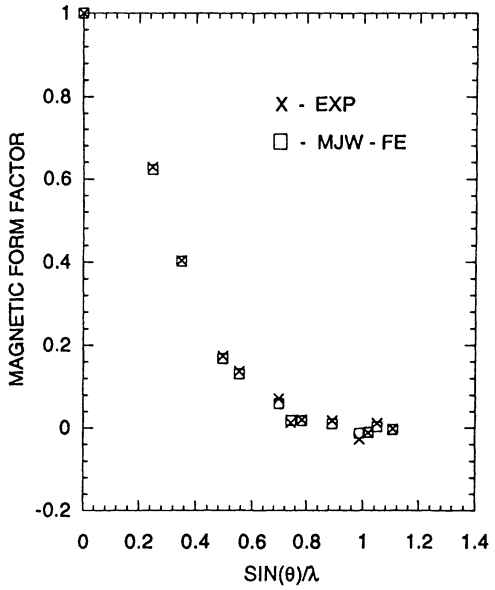


FIG. 4. Comparison of theoretical and experimental form factors normalized to unity at (000) for iron (λ in Å). Theory result based on MJW local density bands; experimental results from Shull and Yamada (Ref. 13).

10% of the form factor and, in some cases (e.g., 400,420,440), in excess of 20%. This is hardly negligible and needs to be borne in mind when evaluating calculations that include only the spin part. It is interesting to reevaluate the calculations of Wang and Callaway[7] in this light. They present results using both Kohn-Sham-Gaspar (KSG) and von Barth–Hedin (VBH) potentials. The inclusion of an orbital contribution into their calculations (with suitable scaling so that the form factor is normalized to unity at 000) actually improves the fit to

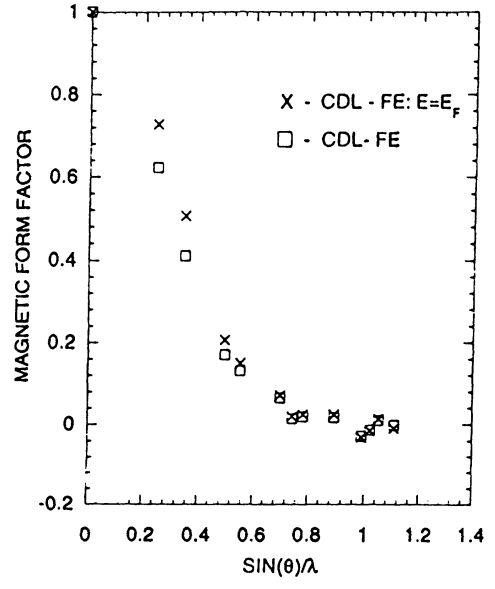


FIG. 5. Comparison of results for theoretical form factor normalized to unity at (000) and calculated with and without energy-dependent radial functions (λ in Å).

TABLE II. Spin, orbital, and total magnetic form factor for nickel calculated from CLD bands.

| *hkl* | Spin | Orbital | Total |
|---|---|---|---|
| 000 | 0.904 | 0.096 | 1.000 |
| 200 | 0.614 | 0.070 | 0.684 |
| 220 | 0.411 | 0.054 | 0.465 |
| 400 | 0.120 | 0.035 | 0.155 |
| 420 | 0.112 | 0.029 | 0.141 |
| 440 | 0.052 | 0.018 | 0.070 |
| 600 | −0.034 | 0.015 | −0.019 |
| 620 | −0.019 | 0.013 | −0.006 |
| 640 | 0.000 | 0.009 | 0.009 |
| 800 | −0.058 | 0.006 | −0.052 |

experimental data (for both KSG and VBH) for values of $\sin\theta/\lambda$ smaller than $\sim 0.7$ Å$^{-1}$, giving agreement comparable to that obtained using CLD bands and considerably better than the MJW (local-density-approximation) values. For large values of $\sin\theta/\lambda$, the fit to the small values of the form factor deteriorates considerably, however.

## CONCLUSIONS

Expressions for the form factor for itinerant-electron systems have been derived and numerically evaluated for ferromagnetic nickel and iron. To our knowledge these are the first calculations based on itinerant-electron theory for *d*-band magnets which include both the spin and orbital parts of the form factor. Since the spin-orbit interaction was included in an approximate way, no attempt was made to find the "best fit" to the data. The results of these calculations do, however, indicate that itinerant-electron theory can provide reasonable predictions of the form factor for both nickel and iron.

At this stage the agreement is not as good as can be obtained from the "best fits" to the atomic model. This work gives some indication of why such good agreement can be obtained from the atomic model. Because of the equivalence of the dominant terms in the itinerant and atomic expressions, if an atomic configuration can be found which yields radial functions similar to those obtained from band theory, then, obviously, good agree-

TABLE III. Spin, orbital, and total magnetic form factor for nickel calculated from MJW local density bands.

| *hkl* | Spin | Orbital | Total |
|---|---|---|---|
| 000 | 0.904 | 0.096 | 1.000 |
| 200 | 0.533 | 0.072 | 0.605 |
| 220 | 0.345 | 0.055 | 0.400 |
| 400 | 0.102 | 0.034 | 0.136 |
| 420 | 0.090 | 0.028 | 0.118 |
| 440 | 0.042 | 0.017 | 0.059 |
| 600 | −0.036 | 0.014 | −0.022 |
| 620 | −0.020 | 0.012 | −0.008 |
| 640 | −0.003 | 0.008 | 0.005 |
| 800 | −0.057 | 0.006 | −0.051 |

ment can be obtained by varying the parameters which appear in the atomistic expression. These parameters cannot be obtained within the framework of the atomic model and may, in fact, have no real meaning at the fundamental level.

A better test of the itinerant theory requires results from a self-consistent spin-polarized calculation which includes the spin-orbit interaction. Local density theory, however, may not be the way to proceed, particularly for nickel. The results obtained for nickel based on local density bands (Fig. 2) are not in good agreement with experiment. This could be caused by a number of factors, including the approximate way the spin-orbit interaction was incorporated into the theory. This is an important point, however, since the form factor is a ground-state property for which local density is expected to yield reasonable results.

The main purpose of this paper has been to evaluate the relative contributions made by the spin and orbital parts to the total form factor. Given that the orbital part has been shown to contribute $\sim 10\%$ and, in some case, in excess of $\sim 20\%$, it can hardly be neglected. It would be interesting to develop a fully relativistic calculation for the form factor based on the type of development used, for example, by Fritsche, Noffke, and Eckardt[14] or Krutzen and Springelkamp[15] for the $g$ factor.


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## APPENDIX

The derivation of expression relevant to the orbital part of the form factor is given in this appendix. For $s$-$d$ and $d$-$d$ symmetry terms, the $\mathbf{D}^{\sigma}_{\mu\nu}$ are clearly real. Then, using the result in Eq. (35),

$$\hat{\mathbf{K}}\times\mathbf{D}^{\sigma}_{\mu,\nu}(\mathbf{K},E)=\hat{\mathbf{K}}\times\tfrac{1}{2}[\mathbf{D}^{\sigma}_{\mu,\nu}(\mathbf{K},E)-\mathbf{D}^{\sigma}_{\nu,\mu}(\mathbf{K},E)]\ , \tag{A1}$$

where

$$\mathbf{D}^{\sigma}_{\mu,\nu}(\mathbf{K},E)=i^{l_\nu-l_\mu-1}\int e^{i\mathbf{K}\cdot\mathbf{r}}\phi^{\sigma}_{\mu}(r,E)\nabla\phi^{\sigma}_{\nu}(r,E)d^3r\ . \tag{A2}$$

Define

$$\tilde{R}^{\sigma}_{l_\mu}(r,E)=\frac{R^{\sigma}_{l_\mu}(r,E)}{r^{l}}\ , \tag{A3}$$

$$\tilde{Y}_{\mu}(\hat{\mathbf{r}})=r^{l}Y_{\mu}(\hat{\mathbf{r}})\ . \tag{A4}$$

Then

$$\begin{aligned}
&\int e^{i\mathbf{K}\cdot\mathbf{r}}[\phi^{\sigma}_{\mu}(r,E)\nabla\phi^{\sigma}_{\nu}(r,E)-\phi^{\sigma}_{\nu}(r,E)\nabla\phi^{\sigma}_{\mu}(r,E)]d^3r\\
&\quad=\int e^{i\mathbf{K}\cdot\mathbf{r}}\tilde{Y}_{\mu}(\hat{\mathbf{r}})\tilde{Y}_{\nu}(\hat{\mathbf{r}})[\tilde{R}^{\sigma}_{l_\mu}(r,E)\nabla\tilde{R}^{\sigma}_{l_\mu}(r,E)-\tilde{R}^{\sigma}_{l_\mu}(r,E)\nabla\tilde{R}^{\sigma}_{l_\mu}(r,E)]d^3r\\
&\qquad+\int e^{i\mathbf{K}\cdot\mathbf{r}}\tilde{R}^{\sigma}_{l_\mu}(r,E)\tilde{R}^{\sigma}_{l_\mu}(r,E)[\tilde{Y}_{\mu}(\hat{\mathbf{r}})\nabla\tilde{Y}_{\nu}(\hat{\mathbf{r}})-\tilde{Y}_{\nu}(\hat{\mathbf{r}})\nabla\tilde{Y}_{\mu}(\hat{\mathbf{r}})]d^3r\\
&\qquad\equiv 2i^{(l_\mu+1-l_\nu)}[\mathbf{I}^{\sigma}_{1}(\mathbf{K},E,\mu,\nu)+\mathbf{I}^{\sigma}_{2}(\mathbf{K},E,\mu,\nu)]\\
&\qquad=2i^{(l_\mu+1-l_\nu)}\mathbf{D}^{\sigma}_{\mu\nu}(\mathbf{K},E)\ .
\end{aligned} \tag{A5}$$

By making use of Eq. (21), it is straightforward to show that

$$\mathbf{I}^{\sigma}_{1}(\mathbf{K},E,\mu,\nu)=4\pi\sum_{\eta}\langle j'_{l_\eta}(K,E)\rangle^{\sigma}_{l_\mu,l_\nu}\mathbf{h}^{\eta,\sigma}_{\mu,\nu}Y_{\eta}(\hat{\mathbf{r}})\ , \tag{A6}$$

where

$$\langle j'_{l_\eta}(K,E)\rangle^{\sigma}_{l_\mu,l_\nu}=\int r^3 j_{l_\eta}(Kr)\left[\tilde{R}^{\sigma}_{l_\mu}(r,E)\frac{d\tilde{R}^{\sigma}_{l_\nu}(r,E)}{dr}-\tilde{R}^{\sigma}_{l_\nu}(r,E)\frac{d\tilde{R}^{\sigma}_{l_\nu}(r,E)}{dr}\right]dr \tag{A7}$$

and

$$h^{\eta,\sigma}_{\mu,\nu}=(-1)^{(l_\nu+l_\eta-l_\mu-1)/2}\sum_{\gamma}C^{\gamma}_{l,m_\alpha;\nu}C^{\gamma}_{\eta,\mu}\ . \tag{A8}$$

The $C^{\eta}_{\mu\nu}$ are Clebsch-Gordan coefficients.

A similar expression can be derived by $\mathbf{I}^{\sigma}_{2}$. This expression is based on the identity

$$\nabla_{\alpha}\tilde{Y}_{l,m}(\hat{\mathbf{r}})=\left[\frac{4\pi}{3}\right]^{1/2}(2l+1)\sum_{m}C^{l-1,M}_{1,m_\alpha;lm}\tilde{Y}_{l-1,M}\ . \tag{A9}$$

Then

$$\mathbf{I}^{\sigma}_{2}(\mathbf{K},E,\mu,\nu)=4\pi\left[\frac{4\pi}{3}\right]^{1/2}K\sum_{\eta}\frac{1}{(2l_\eta+1)}\langle J_{l_\eta}(K,E)\rangle^{\sigma}_{l_\mu,l_\nu}\mathbf{H}^{\eta,\sigma}_{\mu,\nu}Y_{\eta}(\hat{\mathbf{K}})\ , \tag{A10}$$

where

$$(\mathbf{H}^{\eta,\sigma}_{\mu,\nu})_{\alpha}=\tfrac{1}{2}[(2l_{\nu}+1)h^{\eta,\sigma}_{\mu,\nu}-(2l_{\mu}+1)h^{\eta,\sigma}_{\nu,\mu}] \tag{A11}$$

and

$$\langle J_{l_{\eta}}(K,E)\rangle^{\sigma}_{l_{\mu},l_{\nu}}=\int r^2 R^{\sigma}_{l_{\mu}}(r,E)R^{\sigma}_{l_{\nu}}(r,E)[j_{l_{\eta}+1}(Kr)+j_{l_{\eta}-1}(Kr)]dr\ . \tag{A12}$$

The result given in Eq. (37) follows from the definition

$$\mathbf{S}^{\sigma}_{\eta}(\mathbf{K},E)=\sum_{\mu,\nu}\mathbf{H}^{\eta,\sigma}_{\mu,\nu}\tilde{\rho}^{\sigma}_{\mu,\nu}(E)\ . \tag{A13}$$

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