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# Magnetic susceptibility of ferromagnetic metals: Application to nickel

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Expressions for the transverse and longitudinal magnetic (spin) susceptibility of ferromagnetic metals, which have been derived from density-functional theory in the local-density approximation, are simplified and made practical for computation through a transformation from a Fourier representation to an orbital representation. The method is applied to a calculation of the transverse susceptibility of ferromagnetic nickel at $T=0$. Results are obtained for spin-wave energies and widths using a semiempirical band structure.

## I. INTRODUCTION

The response of a system to a small magnetic perturbation is described by linear-response matrices, the wave vector, and frequency-dependent susceptibilities. If the system is ferromagnetic, it is necessary to consider separately the transverse and longitudinal susceptibilities. The latter is coupled to the charge response, so that the dielectric function is derived from the same calculation as is the longitudinal susceptibility. In a paramagnet, the spin and charge responses are not coupled.

Experimental information concerning the dynamic magnetic susceptibility is obtained from the measurement of the cross section for the magnetic scattering of slow neutrons.[1] Consider the double differential cross section for the scattering of a neutron into solid angle $\Omega$ with energy transfer $E=\hbar\omega$. The momentum transfer in the process will be written as

$$\vec{k}_i-\vec{k}_f=\vec{p}_s=\vec{p}+\vec{K}_s\ ,$$

where $\vec{p}$ is a vector in the Brillouin zone and $\vec{K}_s$ is some reciprocal-lattice vector. The basic result is

$$\frac{d^2\sigma}{d\Omega\,dE}\sim\frac{k_f}{k_i}\,\frac{1}{1-\exp(-\hbar\omega/k_BT)}\times\sum_{\alpha,\beta}\left[\delta_{\alpha\beta}-\frac{p_{s\alpha}p_{s\beta}}{p_s^2}\right]\mathrm{Im}\{[\chi^{\alpha\beta}(\vec{p},\omega)]_{ss}\}\ . \tag{1.1}$$

In this expression $k_f$ ($k_i$) is the magnitude of the final (initial) neutron wave vector, $\alpha,\beta$ designate rectangular components, and $\chi_{ss}$ is the $s$-$s$ element of the dynamic-spin-susceptibility tensor matrix.

We shall be concerned here only with cubic ferromagnets at $T=0$ K. In this case, one has two distinguishable susceptibility quantities. Let the direction of magnetization of the sample define the $z$ axis. Then we have two parallel and one perpendicular susceptibilities $\chi^{xx}=\chi^{yy}$ and $\chi^{zz}$, respectively. However, it is now customary instead to introduce $\chi^{+-}$ and $\chi^{-+}$ (Ref. 2),

$$\chi^{xx}+\chi^{yy}=\tfrac{1}{2}(\chi^{-+}+\chi^{+-})\ . \tag{1.2}$$

In addition, we have the symmetry relations

$$\begin{aligned}\mathrm{Re}\chi^{+-}(p,\omega)&=\mathrm{Re}\chi^{-+}(p,-\omega)\ ,\\ \mathrm{Im}\chi^{+-}(p,\omega)&=-\mathrm{Im}\chi^{-+}(p,-\omega)\ .\end{aligned} \tag{1.3}$$

Therefore, knowledge of one of the objects $\chi^{+-}$ or $\chi^{-+}$ for all frequencies is sufficient to determine the total transverse contribution. When neutron scattering measurements are made with polarized neutrons, it is possible to separate the contributions of $\chi^{+-}$ and $\chi^{-+}$, and this has been accomplished in a recent experiment.[2]

Modern investigations of the dynamic susceptibilities were initiated by a fundamental paper of Izuyama, Kim, and Kubo.[3] These authors applied the random-phase approximation to a one-band Hubbard[4] Hamiltonian. The band was treated in a tight-binding approximation. Their result for the transverse susceptibility $\chi^{+-}$ is

$$\chi^{+-}(\vec{p},\omega)=\frac{\chi^{+-(0)}(\vec{p},\omega)}{1-V\chi^{+-(0)}(\vec{p},\omega)}\ , \tag{1.4}$$

in which $V$ is the electron-interaction parameter of the Hubbard model and $\chi^{(0)}$ is the so-called noninteracting susceptibility. They also obtained a formula for the longitudinal susceptibility in the same approximation. Multiband generalizations of Eq. (1.4) have been given by several authors[5–8]; these frequently involve some restrictions of the interaction or approximations concerning wave functions.

The structure of Eq. (1.4) is quite important. If the denominator should vanish for some real $\omega$ for a given $\vec{p}$, the transverse susceptibility would have a pole. This pole would correspond to a spin-wave excitation. In fact, $\chi^{+-(0)}$ is usually a complex function but the spin-wave energy is given by the energy at which the real part of the denominator vanishes, and the lifetime can be determined from the ratio of the imaginary part to the energy derivative of the real part.

There have been many attempts to improve the treatment of many-body effects beyond the random-phase approximation. We do not discuss these here. Our attention is devoted to attempts to take the actual, complicated band structures of real transition metals into account. A major investigation of this type was reported in 1970 by Lowde and Windsor.[9] They compared neutron scattering measurements of the magnetic response function of nickel over a temperature range from $T_c/2$ to $2T_c$ with results of calculations in which Eq. (1.4) is used in combination with a calculation of $\chi^{+-(0)}$ based on a simple tight-binding model of a $d$ band. Lowde and Windsor presented contour plots of the theoretical and experimental susceptibilities in an $\omega,p$ plane at four temperatures, which, although noisy, can be interpreted as showing a basic, qualitative agreement in shape and in temperature dependence.

The work of Lowde and Windsor was extended in significant respects in a series of calculations culminating in the paper of Cooke, Lynn, and Davis.[10] These authors considered more realistic band models of both nickel and iron. The bands were derived from calculations for the paramagnetic state of the material through the introduction of an exchange splitting which gives the correct magnetic moment. This splitting defines the effective electron-electron interaction in a multiband generalization of Eq. (1.4). Spin-wave dispersion curves were obtained which are in good agreement with experiment for both metals. In addition, the existence of an optical spin-wave mode for certain values of $p$ was predicted, also in agreement with experiment.

The work we are reporting here is based on density-functional theory in the local-density approximation. It was shown in previous work[11] that this approach leads to a multiband expression for the transverse susceptibility which has a structure similar to that of Eq. (1.4), but with a different definition of the effective interaction which is determined by the exchange-correlation potential and the magnetization density in the underlying band calculation. An expression for the longitudinal susceptibility has also been derived under the same approximation.[12] It is possible to obtain an explicit formula for the spin-wave stiffness coefficient from the density-functional result just mentioned for the susceptibility. Rather good numerical[13] results for this quantity have been obtained in the case of nickel. Our objective here is to study spin-wave lifetimes as well as energies. In this paper we will present the formalism which makes possible practical calculations of both transverse and longitudinal susceptibilities on the basis of these formulas. Numerical results will be presented for the transverse susceptibility of nickel with the use of a semiempirical band structure. In future work we hope to study the dependence of the susceptibility on the underlying band structure, and also to obtain numerical results for the wave-vector and frequency dependence of the longitudinal susceptibility.

## II. THEORY: TRANSVERSE SUSCEPTIBILITY

The transverse wave-vector and frequency-dependent spin-susceptibility matrix for a ferromagnet is given, within the local spin-density approximation, by the expression[11]

$$\chi=\chi_0(1-\Lambda\chi_0)^{-1} . \tag{2.1}$$

In this equation $\chi_0$ is the non-self-consistent susceptibility matrix (we do not write the indices $+-$, etc., from this point) and $\Lambda$ is a matrix representing $V_f(\vec{r})/M_0(\vec{r})$, $M_0$ being the (dimensionless) magnetization density per atom, and $V_f(\vec{r})$, the interaction leading to ferromagnetism, which is self-consistently determined in the course of a band calculation.[14] The basis vectors for the representation discussed are plane waves whose wave vectors are reciprocal-lattice vectors. An expression has also been derived for the longitudinal spin susceptibility within the same approximations[12]: It will be discussed in Sec. III.

Equation (1) should be compared with the result obtained in the random-phase approximation to the Hubbard model. The latter is of similar appearance, except that the matrix character of the quantities is neglected, $\Lambda$ is replaced by the interaction constant, and $\chi_0$ does not contain form factors. The matrix inversion indicated in Eq. (2.1) is the principal topic of concern here (a similar situation arises in regard to the longitudinal susceptibility as will be discussed subsequently). In previous work, the matrix to be inverted has been approximated as one of modest size ($15\times15$ in a particular application to nickel[15]). We believe, however, that the convergence of this type of calculation as the number of reciprocal-lattice vectors is increased is governed primarily by the spatial extent of the wave functions of the magnetically active electrons. Since the $3d$-electron wave functions in iron and nickel are quite compact, it is likely that the matrices must have dimensions of several thousands in order to obtain converged results.

Therefore, it seems improbable that attempts at direct evaluation of Eq. (1) can yield conclusive results. We are led to consider approximations which enable an analytic solution to the matrix inversion problem associated with (1).

The essential approximations are those which lead to a sort of factorization of $\chi_0$. To see what is involved, we recall the general expression for $\chi_0$,

$$[\chi_0(p,\omega)]_{st}=-\frac{1}{N}\sum_{l,n,\vec{k}}\frac{f_n(\vec{k})-f_l(\vec{k}+\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\vec{p})+\omega+i\eta}\langle n,\vec{k}\,|\,e^{-i\vec{p}_s\cdot\vec{r}}s_+\,|\,l,\vec{k}+\vec{p}\rangle\langle l,\vec{k}+\vec{p}\,|\,e^{i\vec{p}_t\cdot\vec{r}}s_-\,|\,n,\vec{k}\rangle . \tag{2.2}$$

The following conventions and notations are used in this paper: $|\,n,\vec{k}\rangle$ is a Bloch state belonging to band $n$ and wave vector $\vec{k}$; its energy is $E_n(\vec{k})$ and its occupation probability is $f_n(\vec{k})$ (a Fermi function). The operators $s_+$ ($s_-$) raise or lower the electron-spin state. The band index implicitly specifies the spin (in the assumed absence of spin-orbit coupling). Odd-numbered bands are occupied by electrons of spin parallel to the majority (↑), even-numbered bands belong to minority (↓) spin electrons.

Let us consider the explicit expression for the matrix elements in the situation where, as in our band programs, the electron wave functions are expressed as linear combinations of localized orbitals:

$$\psi_n(\vec{k},\vec{r})=N^{-1/2}\sum_{\nu}e^{i\vec{k}\cdot\vec{R}_\nu}\sum_{i}c_{in}(\vec{k})u_i(\vec{r}-\vec{R}_\nu) , \tag{2.3}$$

in which $N$ is the (large) number of cells in the crystal considered, $\vec{R}_\nu$ is a lattice vector, $u_i(\vec{r}-\vec{R}_\nu)$ is the $i$th orbital basis function, and $c_{in}(\vec{k})$ is a (real) normalized eigenvector component determined by diagonalization of the single-particle Hamiltonian. We denote the matrix element considered simply by $M$,

$$M \equiv \langle n,\vec{k}\,|\,e^{-i\vec{p}_s\cdot\vec{r}}s_+\,|\,l,\vec{k}+\vec{p}\rangle = \sum_{\mu} e^{i\vec{k}\cdot\vec{R}_\mu}\sum_{i,j} c_{in}(\vec{k})c_{jl}(\vec{k}+\vec{p})\int u_i(\vec{r})e^{-i\vec{p}_s\cdot\vec{r}}u_j(\vec{r}-\vec{R}_\mu)d^3r \ . \tag{2.4}$$

We have dropped $s_+$, as it is sufficient to require henceforth that $l$ refers to down spin and $n$ to up spin.

The essential conditions for the simplification of the matrix inversion were already discussed by Sokoloff.[5] With reference to Eq. (2.4), it is necessary that the matrix element should be expressible as a sum of products of $\vec{k}$ independent factors which depend on the reciprocal-lattice vector and functions of $k$ independent of $\vec{K}_s$. In order for this to happen we have to abolish the sum over direct-lattice vectors. We must neglect the overlap of wave functions on different sites in (2.4), which means in turn that the basis functions for the band calculation are treated as independent of $k$. This assumption, which is, strictly speaking, in violation of Bloch's theorem, is in fact incorporated in other calculations of transition-metal susceptibilities. The approximation of neglecting overlap in the susceptibility calculation is not unreasonable for $d$ bands where the overlap is in fact small. We believe that it is consistent with this point of view not to include $s$-$d$ hybridization either.

It may be supposed that once we have neglected wave-function overlap, we have retreated to the form-factor approximation in which the eigenvector coefficients $c_{in}$ are replaced by $\delta_{in}$, and only diagonal cases of the integral $(i=j)$ are retained so that the entire susceptibility matrix element is the product of a form factor depending on $\vec{p}_s$ and an energy-dependent factor. This approximation is rather frequently employed, but we will see that it is not necessary. We can retain much of the structure of the solid-state wave functions, in particular the $\vec{k}$-dependent mixing of the different angular symmetries of the dominant "central-cell" $d$-state wave functions. So in fact, we ought to be able to investigate the reliability of the form-factor approximations.

We express our reciprocal-lattice—vector integrals as

$$I_{ij}(\vec{p}_s) = \int u_i(\vec{r})e^{-i\vec{p}_s\cdot\vec{r}}u_j(\vec{r})d^3r \ , \tag{2.5}$$

and then write

$$M = \sum_{i,j} c_{ni}(\vec{k})I_{ij}(\vec{p}_s)c_{lj}(\vec{k}+\vec{p}) \ . \tag{2.6}$$

Additional approximations are not required in the formal theory beyond this point. We will discuss the evaluation of the $I_{ij}$ subsequently.

We can now write the non-self-consistent susceptibility matrix elements in the form

$$[\chi_0(p,\omega)]_{st} = \sum_{\substack{i,j,\\ i',j'}} I_{ij}(\vec{p}_s)I_{i'j'}(-\vec{p}_t)\gamma_{ij,i'j'}(\vec{p},\omega) \ , \tag{2.7}$$

in which

$$\gamma_{ij,i'j'}(\vec{p},\omega) = -\frac{1}{N}\sum_{l,n,\vec{k}} \frac{[f_n(\vec{k})-f_l(\vec{k}+\vec{p})]c_{ni}(\vec{k})c_{lj}(\vec{k}+\vec{p})c_{ni'}(\vec{k})c_{lj'}(\vec{k}+\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\vec{p})+\omega+i\eta} \ . \tag{2.8}$$

It is important that the $\gamma$'s do not depend on the reciprocal-lattice vectors.

The self-consistent susceptibility is now considered following the method of Sinha, Gupta, and Price.[16] Recall that $\Lambda$ in (1) is the Fourier transform of a spatially periodic function. Let us denote the matrix in the denominator by $J$. Its matrix elements on the reciprocal-lattice—vector basis are

$$J_{st} = \delta_{st} - \sum_{u}\sum_{\substack{i,j,\\ i',j'}} \Lambda(\vec{K}_s-\vec{K}_u)I_{ij}(\vec{p}_u)\gamma_{ij,i'j'}I_{i'j'}(-\vec{p}_t) \ . \tag{2.9}$$

We require $J^{-1}$. This can be written in the form

$$(J^{-1})_{st} = \delta_{st} + \sum_{u}\sum_{\substack{i,j,\\ i',j'}} \Lambda(\vec{K}_s-\vec{K}_u)I_{ij}(\vec{p}_u)S_{ij,i'j'}I_{i'j'}(-\vec{p}_t) \ , \tag{2.10}$$

in which a new matrix $S$, whose indices refer to the orbital basis, has been introduced. Consideration of the product $J^{-1}J$ leads via a straightforward calculation involving only some relabeling of indices to

$$S_{ij,i'j'} = \sum_{l,m} \gamma_{ij,lm}[(I-X)^{-1}]_{lm,i'j'} \ , \tag{2.11}$$

in which all indices refer to the orbital basis and

$$X_{ij,i'j'} = \sum_{\substack{s,t,\\ l,m}} I_{ij}(-\vec{p}_s)\Lambda(\vec{K}_s-\vec{K}_t)I_{lm}(\vec{p}_t)\gamma_{lm,i'j'} \ . \tag{2.12}$$

At this point, we have exchanged the problem of inverting a very large matrix on the reciprocal-lattice basis for the problem of inverting a smaller matrix $(I-X)$ on the orbital basis. This is the essential step of our procedure. Finally, we have to form the product $\chi_0 J^{-1}$. This leads to

$$(\chi)_{st} = (\chi_0)_{st} + \sum_{\substack{i,j,\\ i',j'}} I_{ij}(\vec{p}_s)\gamma_{ij,i'j'}(\vec{p},\omega)\sum_{\substack{l,m,\\ l',m'}}\left[\sum_{u,w} I_{i'j'}(-\vec{p}_u)\Lambda(\vec{K}_u-\vec{K}_w)I_{lm}(\vec{p}_w)\right]S_{lm,l'm'}(\vec{p},\omega)I_{l'm'}(-\vec{p}_t) \ . \tag{2.13}$$

It will be observed that the summations over reciprocal-lattice vectors occur in Eqs. (2.12) and (2.13) in forming one type of quantity: a matrix in the band indices

$$B_{i'j',lm}=\sum_{u,w}I_{i'j'}(-\vec{p}_u)\Lambda(\vec{K}_u-\vec{K}_w)I_{lm}(\vec{p}_w)\ , \qquad (2.14)$$

which is independent of energy. This sum, although lengthy, needs to be done only once for each wave vector $\vec{p}$ considered.

Equation (2.13) can be written in the following way analogous to the formula Eq. (2.7) for $\chi_0$:

$$\chi_{st}(\vec{p},\omega)=\sum_{i,j,i',j'}I_{ij}(\vec{p}_s)\Gamma_{ij,i'j'}I_{i'j'}(-\vec{p}_t)\ , \qquad (2.15)$$

in which

$$\Gamma_{ij,i'j'}=\gamma_{ij,i'j'}+\sum_{\substack{l,m,\\ l',m'}}\gamma_{ij,lm}(\vec{p}\omega)B_{lm,l'm'}(\vec{p}\,)S_{l',m',i',j'}(\vec{p},\omega)\ . \qquad (2.16)$$

It is easily verified that the matrices $\gamma$ and $B$ are symmetric with respect to the interchange of orbital pair indices. Equation (2.16) can be rewritten again in the form, after some algebra,

$$\Gamma_{ij,i'j'}=\sum_{l,m}\gamma_{ij,lm}[(I-X)^{-1}]_{lm,i'j'}\ , \qquad (2.17)$$

in which $X=B\gamma$ is the matrix defined in Eq. (2.12). The quantity $\Gamma$ may be regarded as the susceptibility, calculated on the orbital basis.

It should be observed that the matrices involved in Eq. (17) have dimension $n^2\times n^2$, where $n$ is the number of bands considered (five in the present case), rather than $n\times n$ as might initially be expected. Reduction to $n\times n$ form will occur only if off-diagonal terms with regard to the band indices in the form factors $I$ and the reduced susceptibilities $\gamma$ are disregarded, i.e., under the approximation

$$I_{ij}=I_i\delta_{ij},\ \ \gamma_{ij,i'j'}=\gamma_{ii'}\delta_{ij}\delta_{i'j'}\ .$$

The energies and widths of spin waves are found by considering the determinant of the matrix $I-X$ in Eq. (2.17). Let $D(E)$ denote this complex function,

$$D(\vec{p},\omega)=\det(I-X)\ . \qquad (2.18)$$

Then the spin-wave energies are found from the equation

$$\mathrm{Re}[\vec{D}(p,E)]=0\ , \qquad (2.19)$$

which determines the dispersion relation $E(\vec{p}\,)$. There may, of course, be more than one solution of this equation in which case there is a possibility of an optical spin-wave mode. If $E_0$ is an energy at which Eq. (2.19) holds, we find on expansion of $D(E)$ near $E_0$ that the spin-wave width $\Gamma$ is given by

$$\Gamma=2\frac{\mathrm{Im}D(E_0)}{\mathrm{Re}D'(E_0)}\ , \qquad (2.20)$$

in which $D'=dD/dE$. The susceptibility then has a resonant denominator

$$\frac{1}{(E-E_0)^2+\Gamma^2/4}\ ,$$

and the spin-wave peak (in energy for given $p$) has the shape of an asymmetric Lorentzian.

## III. LONGITUDINAL SUSCEPTIBILITY

The local-density (approximation to density-functional theory) result for the longitudinal susceptibility was given in Ref. 12. The coupling of spin- and charge-response functions leads to an even more formidable result for this quantity then for the transverse susceptibility. We will show below how under the same approximations described in the preceding section expressions are obtained for the longitudinal-response functions which are reasonably practical to evaluate.

We begin with Eq. (2.31) of Ref. (12) in which the longitudinal-response functions are given formally by an expression which is a generalization of Eq. (2.1) to the longitudinal case

$$\begin{bmatrix}\chi_{\rho\rho} & \chi_{\rho\sigma}\\ \chi_{\sigma\rho} & \chi_{\sigma\sigma}\end{bmatrix}=\begin{bmatrix}1+(\Gamma_\uparrow+\Gamma_\downarrow)a+(\Gamma_\uparrow-\Gamma_\downarrow)b & -(\Gamma_\uparrow-\Gamma_\downarrow)c-(\Gamma_\uparrow+\Gamma_\downarrow)b\\ -(\Gamma_\uparrow-\Gamma_\downarrow)a-(\Gamma_\uparrow+\Gamma_\downarrow)b & 1+(\Gamma_\uparrow+\Gamma_\downarrow)c-(\Gamma_\uparrow-\Gamma_\downarrow)b\end{bmatrix}^{-1}\begin{bmatrix}-(\Gamma_\uparrow+\Gamma_\downarrow) & \Gamma_\uparrow-\Gamma_\downarrow\\ \Gamma_\uparrow-\Gamma_\downarrow & \Gamma_\uparrow+\Gamma_\downarrow\end{bmatrix}\ . \qquad (3.1)$$

In this equation the $\chi$'s are (infinite) matrices on the reciprocal-lattice-vector basis representing the charge and spin response; thus, for example, $\chi_{\sigma\sigma}$ is the usual longitudinal susceptibility. The $\Gamma_\sigma$ ($\sigma=\uparrow,\downarrow$) are matrices whose elements on the basis are

$$\Gamma_{st,\sigma}=\frac{-1}{N\Omega}\sum_{l,n,\vec{k}}\frac{f_n(\vec{k}\,)-f_l(\vec{k}+\vec{p}\,)}{E_n(\vec{k}\,)-E_l(\vec{k}+\vec{p}\,)+\omega+i\eta}\langle n,\vec{k}\,|\,e^{-i\vec{p}_s\cdot\vec{r}}\,|\,l,\vec{k}+\vec{p}\,\rangle\langle l,\vec{k}+\vec{p}\,|\,e^{i\vec{p}_t\cdot\vec{r}}\,|\,n,\vec{k}\,\rangle\ . \qquad (3.2)$$

The symbols in (3.2) have the same meaning as in (2.2). However, it is to be noted that in this case the bands $n$ and $l$ correspond to the same spin ($\sigma$) whereas opposite spins were involved in (2.2). The quantities $a$, $b$, and $c$ are matrices representing derivatives of the spin-dependent exchange potentials. The expressions for the elements are given below:

$$a_{st}=V_c(\vec{p}_s)\delta_{st}+\tfrac{1}{4}\sum_{\sigma,\sigma'}\left[\vec{K}_s\left|\frac{\partial V_{xc\sigma}}{\partial\rho_{\sigma'}}\right|\vec{K}_t\right]\ , \qquad (3.3)$$

$$b_{st}=\tfrac{1}{4}\sum_{\sigma,\sigma'}\sigma\left[\vec{K}_s\left|\frac{\partial V_{xc\sigma}}{\partial\rho_{\sigma'}}\right|\vec{K}_t\right]\ , \qquad (3.4)$$

and

$$c_{st}=\tfrac{1}{4}\sum_{\sigma,\sigma'}(\sigma\sigma')\left[\vec{\mathbf{K}}_s\left|\frac{\partial V_{xc\sigma}}{\partial\rho_{\sigma'}}\right|\vec{\mathbf{K}}_t\right] , \tag{3.5}$$

and

$$V_c(p_s)=8\pi/\vec{\mathrm{p}}_s^{\,2} , \tag{3.6}$$

$$\left[\vec{\mathbf{K}}_s\left|\frac{\partial V_{xc\sigma}}{\partial\rho_{\sigma'}}\right|\vec{\mathbf{K}}_t\right]=\frac{1}{\Omega}\int_\Omega e^{-i(\vec{\mathrm{K}}_s-\vec{\mathrm{K}}_t)\cdot\vec{\mathrm{r}}}\frac{\partial V_{xc\sigma}}{\partial\rho_{\sigma'}}d^3r . \tag{3.7}$$

The integral is carried out within a unit cell of volume $\Omega$.

Equation (3.1) requires the inverse of a matrix whose elements are $\infty$ dimensional blocks. However, the situation can be alleviated if we follow the methods of the preceding section. In order to simplify somewhat formidable problems of notation, we find it useful to suppress explicit use of indices as much as possible through the use of matrix notation. Thus we rewrite $\Gamma_{st,\sigma}$ as given by (3.2) with the use of the approximation leading to Eq. (2.7) as

$$\Gamma_\sigma=I^T\gamma_\sigma I , \tag{3.8}$$

where $I$ $[I_{ij}(p_s)]$ is now a matrix whose rows are labeled by (pairs of) orbital indices and whose columns are labeled by reciprocal-lattice vectors. $I^T$ is the transpose of the matrix, while $\gamma_\sigma(\gamma_{ij,i'j'})$ is a square matrix whose elements refer to pairs of orbital indices. In addition,

$$g=\gamma_\uparrow+\gamma_\downarrow , \tag{3.9a}$$

$$h=\gamma_\uparrow-\gamma_\uparrow . \tag{3.9b}$$

Then Eq. (3.1) becomes

$$\begin{bmatrix}\chi_{pp} & \chi_{p\sigma}\\ \chi_{\sigma p} & \chi_{c\sigma}\end{bmatrix}=\begin{bmatrix}1+I^TgIa+I^ThIb & -I^ThIc-I^TgIb\\ -I^ThIa-I^TgIb & 1+I^TgIc+I^ThIb\end{bmatrix}^{-1}\begin{bmatrix}-I^TgI & -I^ThI\\ I^ThI & I^TgI\end{bmatrix} . \tag{3.10}$$

Now define

$$L_1=gIa+hIb , \tag{3.11a}$$

$$L_2=hIc+gIb , \tag{3.11b}$$

$$L_3=hIa+gIb , \tag{3.11c}$$

$$L_4=gIc+hIb . \tag{3.11d}$$

We require the inverse matrix

$$\begin{bmatrix}1+I^TL_1 & -I^TL_2\\ -I^TL_3 & 1+I^TL_4\end{bmatrix}^{-1} . \tag{3.12}$$

A formal result for the inverse of a $2\times2$ matrix whose elements are block matrices is

$$\begin{bmatrix}A & B\\ C & D\end{bmatrix}^{-1}=\begin{bmatrix}(A-BD^{-1}C)^{-1} & -(A-BD^{-1}C)^{-1}BD^{-1}\\ -(D-CA^{-1}B)^{-1}CA^{-1} & (D-CA^{-1}B)^{-1}\end{bmatrix} . \tag{3.13}$$

We see by comparison of (3.12) and (3.13) that

$$A=1+I^TL_1 .$$

It is readily verified

$$(1+I^TL_1)^{-1}=1-I^T(1+L_1I^T)^{-1}L_1 . \tag{3.14}$$

The significance of the formal manipulation involved is that whereas $I^TL_1$ is an enormous matrix on the reciprocal-lattice-vector basis, $L_1I^T$ is a matrix of reasonable size on the orbital (pair) basis. Hence it is practical to invert $1+L_1I^T$. We can apply this technique to the more complicated expression which appears in Eq. (3.13).

To simplify the notation slightly, we shall define matrices (in the band indices) $\phi_i$, $i=1,2,3$

$$\phi_i=L_iI^T . \tag{3.15}$$

Consider, for example, the quantity which appears in Eq. (3.13),

$$\begin{aligned}(D-CA^{-1}B)^{-1}&=\{1+I^T[L_4-\phi_3(1+\phi_1)^{-1}L_2]\}^{-1}\\ &=1-I^T[1+\phi_4-\phi_3(1+\phi_1)^{-1}\phi_2]^{-1}[L_4-\phi_3(1+\phi_1)^{-1}L_2] .\end{aligned} \tag{3.16}$$

After considerable algebra of this type, we arrive at expressions for the linear-response functions. The longitudinal spin susceptibility is given by

$$\begin{aligned}\chi_{\sigma\sigma}=I^T\{&[1+\phi_4-\phi_3(1+\phi_1)^{-1}\phi_2]^{-1}\\ &\times[g-\phi_3(1+\phi_1)^{-1}h]\}I .\end{aligned} \tag{3.17}$$

What is particularly to be noted about this formula is that the evaluation of the quantity in curly brackets involves only manipulations of matrices defined on the (relatively) small basis of pairs of orbitals. Even so, the expression is quite complicated. Let us note that in the paramagnetic limit in which

$$\begin{aligned}&\phi_3=0,\ h=0\,,\\ &\chi_{\sigma\sigma}=I^T[(1+\phi_4)^{-1}g]I\\ &\qquad=I^T[(1+gIcI^T)^{-1}g]I\,.\end{aligned}\tag{3.18}$$

This formula is quite analogous to the expression (2.17) for the transverse susceptibility (apart from the suppression of indices). We also have

$$\begin{aligned}\chi_{\rho\rho}=&-I^T\{[1+\phi_1-\phi_2(1+\phi_4)^{-1}\phi_3]^{-1}\\ &\times[g-\phi_2(1+\phi_4)^{-1}h]\}I\,.\end{aligned}\tag{3.19}$$

This expression determines the dielectric function matrix of a ferromagnet. In the paramagnetic limit,

$$\begin{aligned}\chi_{\rho\rho}&=-I^T[(1+\phi_1)^{-1}g]I\\ &=-I^T[(1+gIaI^T)^{-1}g]I\,.\end{aligned}\tag{3.20}$$

Similarly, we find the cross response functions $\chi_{\sigma\rho}$ and $\chi_{\rho\sigma}$,

$$\begin{aligned}\chi_{\sigma\rho}=&I^T\{[1+\phi_4-\phi_3(1+\phi_1)^{-1}\phi_2]^{-1}\\ &\times[h-\phi_3(1+\phi_1)^{-1}g]\}I\,,\end{aligned}\tag{3.21}$$

$$\begin{aligned}\chi_{\rho\sigma}=&-I^T\{[1+\phi_1-\phi_2(1+\phi_4)^{-1}\phi_3]^{-1}\\ &\times[h-\phi_2(1+\phi_4)^{-1}g]\}I\,.\end{aligned}\tag{3.22}$$

These quantities ($\chi_{\sigma\rho}$ and $\chi_{\rho\sigma}$) vanish in the paramagnetic limit.

## IV. TRANSFORMATIONS: A LIMITING CASE

The preceding expressions for response matrices are quite complex, particularly in the case of the longitudinal functions [Eqs. (3.17)–(3.22)]. Some insight into the nature of these expressions is obtained if we transform to a real-space form.

We see from Eqs. (3.11) and (3.15) that the essential quantity to consider has the form

$$\Theta=I\theta I^T\,,\tag{4.1}$$

in which $\theta$ is a matrix in the reciprocal-lattice vectors. We are to regard $\Theta$ as a matrix in the bands (indices correspond to pairs of bands). When indices are restored, we have

$$\Theta_{ij,i'j'}=\sum_{st}I_{ij}(-\vec{p}_s)\theta(\vec{K}_s-\vec{K}_t)I_{i'j'}(\vec{p}_t)\,.\tag{4.2}$$

We insert the definition of the $I_{ij}$ [Eq. (2.5)] and use the fact that $\theta(\vec{K}_s-\vec{K}_t)$ is a Fourier coefficient of some periodic function $\Theta(r)$:

$$\theta(\vec{K}_s-\vec{K}_t)=\frac{1}{N\Omega}\int d^3r\,e^{i(\vec{K}_s-\vec{K}_t)\cdot\vec{r}}\theta(\vec{r})d^3r\,.\tag{4.3}$$

The sums over reciprocal-lattice vectors may then be carried out with the aid of the identity,

$$\sum_s e^{i\vec{K}_s\cdot(\vec{r}-\vec{r}\,')}=\Omega\sum_\mu\delta(\vec{r}-\vec{r}\,'-\vec{R}_\mu)\,,\tag{4.4}$$

in which $\vec{R}_\mu$ is a direct-lattice vector. Then we find

$$\begin{aligned}\Theta_{ij,i'j'}=\Omega\sum_\sigma &e^{i\vec{p}\cdot\vec{R}_\sigma}\\ &\times\int u_i(\vec{r})u_j(\vec{r})\theta(\vec{r})\\ &\qquad\times u_{i'}(\vec{r}-\vec{R}_\sigma)u_{j'}(\vec{r}-\vec{R}_\sigma)d^3r\,.\end{aligned}\tag{4.5}$$

Although Eq. (4.5) contains terms involving orbitals on different sites it would be consistent with our discussion in Secs. II and III to neglect them, and so to arrive at the approximation

$$\Theta_{ij,i'j'}=\int u_i(\vec{r})u_j(\vec{r})\theta(\vec{r})u_{i'}(\vec{r})u_{j'}(\vec{r})d^3r\,.\tag{4.6}$$

We can use this transformation to extract an interesting result in a limiting case. Let us suppose we have only a single band and a single type of orbital. This idealized situation corresponds to the tight-binding "$S$" band often considered in discussions of magnetic materials, although it is not a realistic description of actual magnetic materials. We will consider the uniform ($\omega=0,p\rightarrow0$) longitudinal susceptibility of such a system in the paramagnetic state via Eq. (3.18). The quantity $IcI^T$ reduces to

$$IcI^T\equiv-U=\int u^4(r)c(r)d^3r\tag{4.7}$$

(indices are not written since there is only one orbital). The quantity $U$ replaces in this case the usual interaction constant of the Hubbard model.

It is quite straightforward to show that in the limits considered here, including the neglect of the overlap of wave functions on different sites, that

$$\lim_{\substack{\omega\rightarrow0,\\ p\rightarrow0}}g(p,\omega)=-\int G(E)\frac{df}{dE}dE\,,\tag{4.8a}$$

where $G(E)$ is the density of states and $f$ is the Fermi function. Thus in the additional limit of zero temperature

$$g(0,0)=G(E_F)\,.\tag{4.8b}$$

Thus, we have for the uniform susceptibility matrix,

$$[\chi_{\sigma\sigma}(p=0,\omega=0)]_{st}=I(\vec{K}_s)I(\vec{K}_t)\frac{G(E_F)}{1-UG(E_F)}\,.\tag{4.9}$$

The quantities $I$ are form factors. We have only rederived a standard formula, but we have an explicit expression for the Stoner enhancement. This result is equivalent to that obtained by Vosko and Perdew[17] and by Janak[18] if the corresponding approximation about wave functions are made in their formulas.

Although the algebra is a bit more complicated, a similar result can be obtained in the ferromagnetic case. The uniform limit ($p=0,\omega=0$) of the longitudinal susceptibility is to be discussed. This limit has previously been considered within local spin-density-functional theory by Yamada, Yasui, and Shimizu[19] by a different approach. We shall confine our remarks here to a hypothetical single (but spin-split) band, single orbital, ferromagnet because in this case the matrices $\phi_i$ [Eq. (3.15)] become scalars. It is important to note in the analysis of Eq. (3.17) in this case that the matrix $a$ [Eq. (3.3)] has one diagonal element which tends to infinity. We find when we take the limits and use the generalization of Eq. (4.8) for a ferromagnet

$$[\mathcal{X}_{\sigma\sigma}(p=0,\omega=0)]_{st}=I(K_s)I(\vec{K}_t)\bar{\mathcal{X}}\ , \tag{4.10}$$

in which

$$(\bar{\mathcal{X}})^{-1}=\frac{1}{4}\left[\frac{1}{G_{\uparrow}(E_F)}+\frac{1}{G_{\downarrow}(E_F)}\right]-U\ , \tag{4.11}$$

where $U$ is given by Eq. (4.7). This result is, in form, the same as that obtained in the Stoner-Wohlfarth picture,[20,21] and confirms the designation of $U$ as the Stoner parameter of the present theory.

## V. METHOD OF CALCULATION

It is apparent from Eqs. (2.7) and (2.12) that the calculation of the transverse susceptibility requires computation of three types of quantities: (1) the effective interaction $\Lambda(V_f/M_0)$, (2) the form factors $I_{ij}$, and (3) the reduced susceptibilities $\gamma_{ij,i'j'}$. The ingredients of the longitudinal susceptibility are similar, but are manipulated in a more complex fashion. For reasons discussed in the next section in this paper, the interaction quantities $\Lambda$ are determined by a fit to experimental data. This section is focused on the calculation of the form factors and the reduced susceptibility.

We begin with the form factors. It is convenient and consistent with the band-calculation procedure to suppose that the orbitals $u_i$ have the form of a common $3d$ radial function times an angular function $K_i(\hat{r})$

$$u_i(\vec{r})=R(r)K_i(\hat{r})\ , \tag{5.1}$$

in which $K_i$ is a Kubic harmonic belonging to the $\Gamma_{25'}$ or $\Gamma_{12}$ representations. For future reference, we shall define

$$K_1=Nxy/r^2,\ \ K_2=Nyz/r^2,\ \ K_3=Nxz/r^2\ , \tag{5.2}$$

$$K_4=N(x^2-y^2)/2r^2,\ \ K_5=N(3z^2-r^2)/(2\sqrt{3}r^2)\ ,$$

where the normalization constant $N(15/4\pi)^{1/2}$ is chosen so that

$$\int K_iK_jd\sigma=\delta_{ij} \tag{5.3}$$

($d\sigma$ will denote an element of solid angle). Then consider

$$I_{ij}(\vec{p}\,)=\int u_i(\vec{r}\,)e^{-i\vec{p}\cdot\vec{r}}u_j(\vec{r}\,)d^3r=\sum_l(-i)^l(2l+1)\int R^2(r)j_l(pr)r^2dr\int K_i(\hat{r})P_l(\cos\theta_{\vec{p}\cdot\vec{r}})K_i(\hat{r})d\sigma\ . \tag{5.4}$$

The standard expansion of a plane wave in spherical waves has been employed. The particular choice of angular functions employed here implies that only $l=0,2,4$ can contribute. Hence we have

$$I_{ij}(p)=\delta_{ij}I^{(0)}(p)-5I^{(2)}(p)C_{ij}^{(2)}(\hat{p})+9I^{(4)}(p)C_{ij}^{(4)}(\hat{p})\ , \tag{5.5}$$

in which

$$I^{(l)}(p)=\int R^2(r)j_l(pr)r^2dr \tag{5.6}$$

and

$$C_{ij}^{(l)}(\hat{p})=\int K_i(\hat{r})P_l(\cos\theta_{\vec{p}\cdot\vec{r}})K_j(\hat{r})d\sigma\ . \tag{5.7}$$

The $C_{ij}^{(l)}(\hat{p})$ are polynomials in the components of $\vec{p}$. They can be computed quite straightforwardly by elementary means. One can simply express the Legendre polynomials in terms of the rectangular components of $\vec{p}$ and $\vec{r}$, use the explicit expression for the Kubic harmonics, and do the integrals which are elementary. This process leads to the formulas given in Table I. Rather complicated results involving sums over Clebsch-Gordan coefficients have been obtained by other authors[22] but our results are quite simple. The reader should note that in Table I, the notation $x\equiv p_x/p$, etc., is used.

The radial integral was computed using Clementi wave functions.[23] We have performed the computation for both the $d^8s^2(^3F)$ and $d^9s^1(^3D)$ atomic functions. These functions are combinations of the form

$$R(r)=\sum_j c_jr^2e^{-\zeta_jr}\ , \tag{5.8}$$

involving five exponentials. Therefore, we write

$$I^l(p)=\sum_{jk}c_jc_kI^{(l)}(p,\zeta_j,+\zeta_k)\ , \tag{5.9a}$$

in which (with the aid of a tabulated integral[24])

$$I^{(l)}(p,\zeta)=\int r^6e^{-\zeta r}j_l(pr)dr$$

$$=\tfrac{1}{2}\sqrt{\pi}\left[\frac{p}{2\zeta}\right]^l\frac{\Gamma(l+7)}{\Gamma(l+\frac{3}{2})}\frac{\zeta^5}{(\zeta^2+p^2)^6}$$

$$\times{}_1F_1\left[\frac{l-5}{2},\frac{l-4}{2},l+\frac{3}{2},-\frac{p^2}{\zeta^2}\right]\ . \tag{5.9b}$$

The object ${}_1F_1$ is a hypergeometric function. However, since $l-4$ is an even negative integer or zero, the function is just a polynomial.

The $d^9s^1$ radial wave function is somewhat more extended spatially than the $d^8s^2$ function. Consequently, the $I_{ij}(p)$ are more longer ranged in $p$ for the $d^8s^2$ case than for $d^9s^1$. However, sums over reciprocal-lattice vectors involving $I_{ij}$ [as in Eq. (2.12)] will not converge until $p_s$ becomes large compared to the most important $\zeta$ in the expansion of the wave function (or, alternatively, if $r_0$ locates the peak of the $3d$ function $p_sr_0$ must be large). Consequently, large numbers of lattice vectors must be included for both radial functions, but the convergence is somewhat faster for the $d^9s^1$ functions than for $d^8s^2$.

The most complicated portion of the calculations concerns the evaluation of the quantities $\gamma$, $g$, and $h$ [see Eqs. (2.8) and (3.9)]. Consider the sum over wave vectors in

Eq. (2.8). This has to include all $\vec{k}$ in the zone, but normally one has available results pertaining to the "irreducible wedge," $\frac{1}{48}$th of the zone in the case of a cubic crystal. Then we can express the sum over all $\vec{k}$ as a sum over the $\vec{k}$ in the irreducible wedge combined with a sum over group operations, $\alpha$, which generate all $\vec{k}$ in zone from those in the wedge, $W$. Thus

$$\gamma_{ij,i'j'}=(-)\frac{1}{N}\sum_{\alpha}\sum_{\vec{k}\in W}\sum_{l,n}[f_l(\alpha\vec{k})-f_l(\alpha\vec{k}+\vec{p})]\frac{c_{ni}(\alpha\vec{k})c_{lj}(\alpha\vec{k}+\vec{p})c_{ni'}(\alpha\vec{k})c_{lj'}(\alpha\vec{k}+\vec{p})}{E_n(\alpha\vec{k})-E_n(\alpha\vec{k}+\vec{p})+\omega+i\eta}\ . \tag{5.10}$$

The energies are of course unchanged if $\alpha\vec{k}$ is replaced by $\vec{k}$. This is also true for the Fermi factors which depend on $\vec{k}$ only through the energy. However, the transformation properties of the $c$'s must be considered. In this case we have the rule (when $\vec{k}$ is not a point of degeneracy),

$$c_{nj}(\alpha\vec{k})=\chi_n(\alpha)\sum_l D_{jl}(\alpha)c_{nl}(\vec{k})\ , \tag{5.11}$$

where $D_{jl}$ is an element of the representation matrix describing the transformation of the orbitals $u_j(r)$ under the operations $\alpha$, and $\chi_n(\alpha)$ $(=\pm 1)$ is the character of the operation $\alpha$ in one of the one-dimensional representations of the relevant group (the cubic group in this case).

TABLE I. Polynomials $C_{ij}^{(l)}(\hat{p})$ defined by Eq. (5.7). Note that $C^{(4)}$ is related to $C^{(2)}$ and a polynomial $S$ as shown. In these expressions, $x=p_x/p$, etc.

| | $l=2$ |
|---|---|
| $(i,j)$ | $C_{ij}^{(2)}$ |
| (1,1) | $(x^2+y^2-2z^2)/7$ |
| (1,2) | $3xz/7$ |
| (1,3) | $3yz/7$ |
| (1,4) | $0$ |
| (1,5) | $-2\sqrt{3}xy/7$ |
| (2,2) | $(y^2+z^2-2x^2)/7$ |
| (2,3) | $3xy/7$ |
| (2,4) | $-3yz/7$ |
| (2,5) | $\sqrt{3}yz/7$ |
| (3,3) | $(x^2+z^2-2y^2)/7$ |
| (3,4) | $3xz/7$ |
| (3,5) | $\sqrt{3}xz/7$ |
| (4,4) | $(x^2+y^2-2z^2)/7$ |
| (4,5) | $-\sqrt{3}(x^2-y^2)/7$ |
| (5,5) | $-(x^2+y^2-2z^2)/7$ |
| | $l=4$<br>$C_{ij}^{(4)}=\frac{5}{24}S_{ij}-\frac{5}{2}C_{ij}^{(2)}-\frac{7}{8}\delta_{ij}$ |
| $(i,j)$ | $S_{ij}$ |
| (1,1) | $z^4+5(x^4+y^4)+6z^2(x^2+y^2)+18x^2y^2$ |
| (1,2) | $4xz(x^2+3y^2+z^2)$ |
| (1,3) | $4yz(3x^2+y^2+z^2)$ |
| (1,4) | $4xy(x^2-y^2)$ |
| (1,5) | $-4\sqrt{3}xy(x^2+y^2)$ |
| (2,2) | $x^4+5(y^4+z^4)+6x^2(y^2+z^2)+18y^2z^2$ |
| (2,3) | $4xy(x^2+y^2+3z^2)$ |
| (2,4) | $-4yz(2y^2+z^2)$ |
| (2,5) | $4\sqrt{3}yz^3$ |
| (3,3) | $y^4+5(x^4+z^4)+6y^2(x^2+z^2)+18x^2z^2$ |
| (3,4) | $4xz(2x^2+z^2)$ |
| (3,5) | $4\sqrt{3}xz^3$ |
| (4,4) | $z^4+7(x^4+y^4)+6(x^2y^2+y^2z^2+x^2z^2)$ |
| (4,5) | $-2\sqrt{3}(x^4-y^4)$ |
| (5,5) | $9z^4+3(x^4+y^4)+6(x^2y^2+y^2z^2+x^2z^2)$ |

Equation (5.11) follows from the fundamental rule for the transformation of the Bloch wave function $\phi_n(\vec{k},r)$, providing this is a nondegenerate state,[25]

$$\alpha\psi_n(\vec{k},\vec{r})=\psi_n(\vec{k},\alpha^{-1}\vec{r})=\chi_n(\alpha)\psi_n(\alpha\vec{k}+\vec{r})\ . \tag{5.12}$$

To see that Eq. (5.11) follows from Eq. (5.12), substitute the expansion (2.3) for $\phi(\vec{k},\alpha^{-1}\vec{r})$. The transformation formula for the orbitals $u_j$ is used, which we take to be

$$u_j(\alpha^{-1}\vec{r})=\sum_l D_{lj}(\alpha)u_l(\vec{r})\ . \tag{5.13}$$

Then a straightforward argument leads to (5.11).

Since we are considering in this work only a relatively small number of momentum-transfer vectors $\vec{p}$ along the axes of high symmetry, there are some additional symmetries which can be exploited. Let us consider operators $\beta$ which have the property

$$\beta\vec{p}=\vec{p}$$

(we do not consider cases where $\vec{p}$ is on the surface of the zone). These operators form the group of $\vec{p}$ (denoted $\kappa_p$) which is a subgroup of the cubic group. If there are $n$ operations $\kappa_p$, the integer quantity $m$,

$$m=48/n\ ,$$

gives the number of vectors in the star of $\vec{p}$. We may then select $m$ operators denoted $\theta_i$ $(\theta_1=E)$, such that the cubic group, $G$, can be expressed as a sum of right cosets

$$G=\kappa_p+\kappa_p\theta_2+\cdots+\kappa_p\theta_m\ .$$

Thus any operator $\alpha$ in $g$ can be written in the form

$$\alpha=\beta_i\theta_j\ ,$$

where $\beta_i$ is one of the operators in $\kappa_p$. Then since the band energies are invariant under the operators of $G$,

$$\begin{aligned}E_l(\alpha\vec{k}+\vec{p})&=E_l(\vec{k}+\alpha^{-1}\vec{p})=E_l(\vec{k}+\theta_j^{-1}\beta_j^{-1}\vec{p})\\&=E_l(\vec{k}+\theta_i^{-1}\vec{p})\ .\end{aligned} \tag{5.14}$$

We note that the occupation functions $f$ are functions of the energies only, and therefore also satisfy Eq. (5.14).

All these considerations lead us to rewrite Eq. (5.10) in the form

$$\gamma_{ij,i'j'}=(-)\sum_{n,l}\sum_{\theta}\frac{1}{N}\sum_{\vec{k}\in W}\frac{f_n(\vec{k})-f_l(\vec{k}+\theta^{-1}\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\theta^{-1}\vec{p})+\omega+i\eta}\sum_{\beta}c_{ni}(\beta\theta\vec{k})c_{nj}(\beta\theta\vec{k}+\vec{p})c_{ni'}(\beta\theta\vec{k})c_{lj'}(\beta\theta\vec{k}+\vec{p}) \; . \tag{5.15}$$

The summations are written in the order in which they are actually performed. The summation over $\beta$ includes all the operators in $\kappa_p$, and that over $\theta$ includes the $m$ operators $\theta_m$ defined above. Note that we must use Eq. (5.11) to determine the eigenvectors at the rotated point $\beta\theta\vec{k}$. Then Eq. (5.15) can be rewritten as

$$\gamma_{ij,i'j'}=(-)\sum_{n,l}\sum_{\theta}\frac{1}{N}\sum_{k\in W}\frac{f_n(\vec{k})-f_l(\vec{k}+\theta^{-1}\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\theta^{-1}\vec{p})+\omega+i\eta}$$
$$\times\sum_{\beta}\sum_{\substack{a,a'\\b,b'}}D_{ia}(\beta\theta)D_{jb}(\beta\theta)D_{ia'}(\beta\theta)D_{j'b'}(\beta\theta)c_{na}(\vec{k})c_{lb}(k+\theta^{-1}\vec{p})c_{na'}(\vec{k})c_{lb'}(\vec{k}+\theta^{-1}\vec{p}) \; . \tag{5.16}$$

Since the $D$'s are a (reducible) representation of the cubic group, we can write, for example,

$$D_{ia}(\beta\theta)=\sum_{c}D_{ic}(\beta)D_{ca}(\theta) \; .$$

Then the sum over the members $\beta$ of the group of $p$ can be performed formally by defining

$$U(i,j;i',j';c,d;c',d')=\sum_{\beta}D_{ic}(\beta)D_{jd}(\beta)D_{i'c'}(\beta)D_{j'd'}(\beta) \; . \tag{5.17}$$

We then obtain

$$\gamma_{ij,i'j'}=(-)\sum_{n,l}\sum_{\theta}\frac{1}{N}\sum_{k\in W}\frac{f_n(\vec{k})-f_l(k+\theta^{-1}\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\theta^{-1}\vec{p})+\omega+i\eta}$$
$$\times\sum_{\substack{a,a',\\b,b'}}\sum_{\substack{c,c',\\d,d'}}U(i,j;i',j';c,c';d,d')D_{ca}(\theta)D_{db}(\theta)D_{c'a'}(\theta)D_{c'b'}(\theta)$$
$$\times c_{na}(\vec{k})c_{lb}(\vec{k}+\theta^{-1}\vec{p})c_{na'}(\vec{k})c_{lb'}(\vec{k}+\theta^{-1}\vec{p}) \; . \tag{5.18}$$

Only in the somewhat unphysical extreme tight-binding limit in which the $\vec{k}$ dependence of the eigenvectors is totally neglected and the bands $n$ are placed into one-to-one correspondence with the orbitals $i$ does $\gamma$ simplify in a major way. Then one obtains

$$\gamma_{ij,i'j'}=(-)\delta_{i'i}\delta j'j\sum_{\theta}\frac{m}{N}\sum_{k\in W}\frac{f_i(\vec{k})-f_j(\vec{k}+\theta^{-1}\vec{p})}{E_i(\vec{k})-E_j(\vec{k}+\theta^{-1}\vec{p})+\omega+i\eta} \; . \tag{5.19}$$

Our procedure is to separate Eq. (5.18) into real and imaginary parts using the identity

$$\lim_{\eta\to 0}(x+i\eta)^{-1}=\mathrm{P}\left[\frac{1}{x}\right]=i\pi\delta(x) \; .$$

Note all the other quantities Eq. (5.18) are real.

Let us consider the imaginary part of $\gamma$. The sum over $\vec{k}$ in the wedge is regarded as an integral, and the $\delta$ function gives rise to an integral over surfaces in the wedge of constant energy difference, $\omega$,

$$\frac{1}{N}\sum_{k\in W}\mathrm{Im}\frac{f_n(\vec{k})-f_l(\vec{k}+\theta^{-1}\vec{p})}{E_n(\vec{k})-E_l(\vec{k}+\theta^{-1}\vec{p})+\omega+i\eta}\times\cdots=\frac{\Omega}{(2\pi)^3}\int dS_\omega\frac{f_n(\vec{k})-f_l(\vec{k}+\theta^{-1}\vec{p})}{|\vec{\nabla}_{\vec{k}}[E_n(\vec{k})-E_l(\vec{k}+\theta^{-1}\vec{p})|}\times\cdots \; . \tag{5.20}$$

The integral is evaluated by the linear analytic tetrahedron method.[26–28] The real part of $\gamma$ is then obtained as the Hilbert transform of the imaginary part

$$\mathrm{Re}\gamma_{ij,i'j'}(\vec{p},\omega)=\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\mathrm{Im}\gamma_{ij,i'j'}(\vec{p},\nu)}{\nu-\omega}d\nu \; . \tag{5.21}$$

In spite of the simplifications introduced by our approximations in regard to matrix elements, the numerical calculations are quite lengthy. This is due to the large number (325) of elements $\gamma_{ij,i'j'}$ that have to be determined for each value of $p$ and $\omega$. We have therefore investigated whether the "diagonal" approximation, which sets

$$\gamma_{ij,i'j'} \approx \delta_{ij}\delta_{i'j'}\gamma_{ii,j'j'} , \tag{5.22}$$

would be sufficiently accurate. We calculated $\mathrm{Im}(\chi^{-+}+\chi^{+-})$ for a single value of $\vec{p}$ $[(2\pi/a)(\frac{1}{8},0,0)]$, as a function of $\omega$ both using (5.22), and also including all elements of $\gamma$. The results agreed very well for all energies, indicating that the diagonal approximation is sufficiently accurate.

Our calculations were based on the semiempirical band structure of Weling and Callaway.[29] The reasons for this choice and some of the physical problems involved are discussed at the beginning of the next section. It is to be mentioned here that we considered the lowest six (out of nine) bands, and retain only the $d$-orbital components of the eigenvectors. We used a grid based on 240 points in the irreducible $\frac{1}{48}$th wedge of the Brillouin zone.

## VI. RESULTS AND DISCUSSION

The results reported here are based on the semiempirical band structure determined by Weling and Callaway[29] which is in substantial agreement with both Fermi-surface and photoemission measurements. This choice was made on the belief that the susceptibility would be sensitive to the exchange splitting. The semiempirical band structure is subject to some uncertainties in regard to possible incorporation of final-state effects (in regard to photoemission) but probably gives a reasonably correct account of the exchange splitting. It would, of course, be of interest to repeat the susceptibility calculation using the "first-principles" bands.[30] We hope to report that calculation in the future.

An obvious difficulty connected with the use of semiempirical bands is that the radial $3d$ wave function is not determined. We have therefore used an atomic wave function for the calculation of the form factor. Results have been obtained both with the Hartree-Fock wave functions for the $^3F$ state of the $d^8s^2$ configuration and for the $^2D$ state of the $d^9s^1$ configuration.[23] The form factor based on the $d^8s^2$ function is somewhat more extended in momentum space because the wave function is more compact in position space. However, because we introduce an adjustable constant to represent the interaction this makes little difference to our most important results. Those shown are based on the $d^9s^1$ function.

The most serious problem connected with the use of the semiempirical band structure concerns the quantities $\Lambda$ [see definition following Eq. (2.1)]. The basic assumption here is that there exists some local potential from which the empirical band structure could be derived, but we obviously do not know what it is. For lack of any better option, we replace the matrix $\Lambda_{st}=\Lambda(\vec{K}_s-\vec{K}_t)$ by a multiple of the unit matrix

$$\Lambda_{st}=\lambda\delta_{st} ,$$

where the constant $\lambda$ becomes our single adjustable parameter. A rough estimate of $\lambda$ can be made on the basis of the semiempirical bands if we replace the quantities involved in its definition by suitable averages. Presumably, the averages should be taken with a typical $3d$-electron wave function. Then

$$\lambda=\langle V_f\rangle/\langle M_0(r)\rangle , \tag{6.1}$$

in which $M_0(r)$ is the magnetization density. We have

$$\langle V_f\rangle=-\tfrac{1}{2}\langle\Delta_{\mathrm{ex}}\rangle , \tag{6.2}$$

in which $\Delta_{\mathrm{ex}}$ is the average exchange splitting for which we use 0.3 eV. The average of the magnetization requires us to consider the fact that the $d$ electrons occupy only a small fraction, $f$, of the volume of the unit cell. Then

$$\langle M_0(r)\rangle \approx \int |\psi_{3d}(r)|^2(-\tfrac{1}{2})(\rho_\uparrow-\rho_\downarrow)d^3r$$
$$\approx(-\tfrac{1}{2})\frac{1}{f}(n_\uparrow-n_\downarrow) . \tag{6.3}$$

The factor of $-\frac{1}{2}$ results from the definition of $M$ employed in Ref. 11, and $n_{\uparrow(\downarrow)}$ is the number of majority (minority) spin $3d$ electrons per atom. Thus

$$\lambda\approx f\frac{\Delta_{\mathrm{ex}}}{n_\uparrow-n_\downarrow}=0.5f \tag{6.4}$$

(in units of eV), where we have used $n_\uparrow-n_\downarrow=0.6$. The fraction $f$ is in fact reasonably small as the maximum of the $d$-electron wave functions is well inside the cell. A reasonable guess as to the fraction of the cell volume in which the magnetization density is important is probably in the range of 10–20 %, leading to

$$\lambda\approx0.05-0.10$$

(measured in eV). We determine the value of $\lambda$ by fitting our results as closely as possible to the observed spin-wave spectrum for small $p$. We take this to be given by[31]

$$\omega=Dp^2(1-\beta p^2) , \tag{6.5}$$

where $D$ is the spin-wave stiffness. However, there are some problems. We can make calculations only for values of $p$ corresponding to grid points on which the band structure is calculated. These points may be characterized by integers $(n_1,n_2,n_3)$ so that the vector $\vec{p}$ is given by

$$\vec{p}=\frac{2\pi}{a}\left[\frac{n_1}{12},\frac{n_2}{12},\frac{n_3}{12}\right] .$$

Thus the smallest value of $p$ for which we can make calculations is $\frac{1}{12}$th of the $\Gamma-X$ distance in the zone. As $p$ increases, there are significant departures in theory and in experiment from the form of Eq. (6.5) both in regard to magnitude and in regard to directional anisotropy. We find a substantial degree of agreement with experiment for $\lambda=0.094$ eV.[32]

Our results for the spin-wave energies are shown in Fig. 1, where they are compared with those obtained from the empirical formula, Eq. (6.5), with $D=593$ meV Å$^2$, $\beta=0.68$ Å$^2$ which represents the observed spin-wave spectrum at low temperatures according to the measurements of Mook and Lynn.[31]

The agreement between the calculated and the experimental energies is fairly good, showing that our calculation reproduces the essential features of the spin-wave spectrum. As $p$ increases, the results deviate from the representation of Eq. (6.5) with the larger deviation occurring in the (111) direction. This is qualitatively in accord with

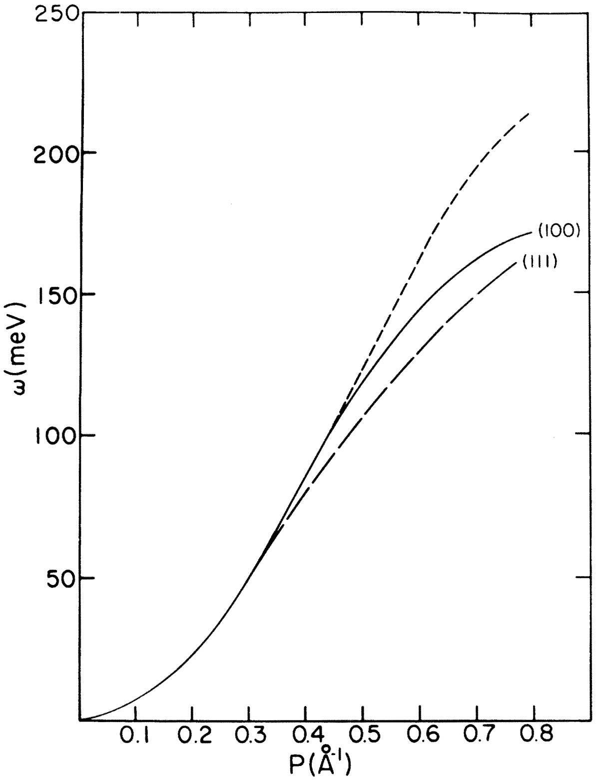

FIG. 1. Calculated spin-wave energies for wave vectors along the [100] and [111] axis. The dashed short curve represents the empirical fit to the experimental spin-wave energies given in Ref. 31.

the results of Mook and Tocchetti.[38] However, their results pertain to room temperature, so direct comparison is not possible.

As the spin-wave energies increase, so does the width. This is apparent from Figs. 2 and 3 in which we show the imaginary part of the $K_s = K_t = 0$ element of the combined transverse susceptibility matrix $(\chi^{-+} + \chi^{+-})$ for five values of $p$ along the [100] axis and three along the [111] axis. The curves flatten and broaden with increasing $p$. Numerical results for the energies and widths are contained in Table II. [The widths were determined from Eq. (2.20).] The experimental results of Ref. 31 cannot be compared directly with our calculation. However, a rough estimate based on Fig. 9 of Ref. 31 indicates reasonable agreement for low energies.

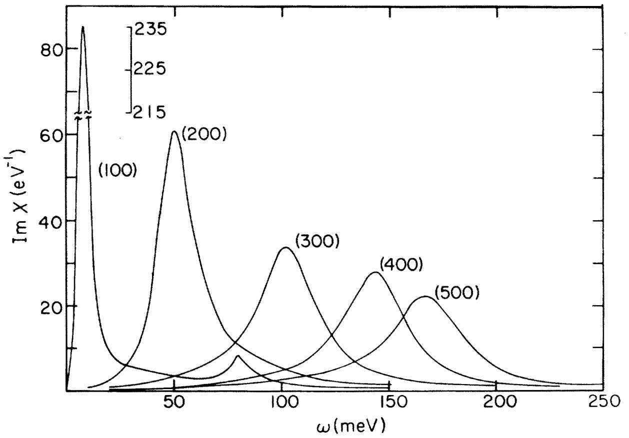

FIG. 2. Imaginary part of the transverse susceptibility $(\chi^{-+} + \chi^{+-})$ for $\vec{p}$ along the [100] axis. Curves are labeled by the value of $\vec{p}$ in units of $2\pi/(12a)$.

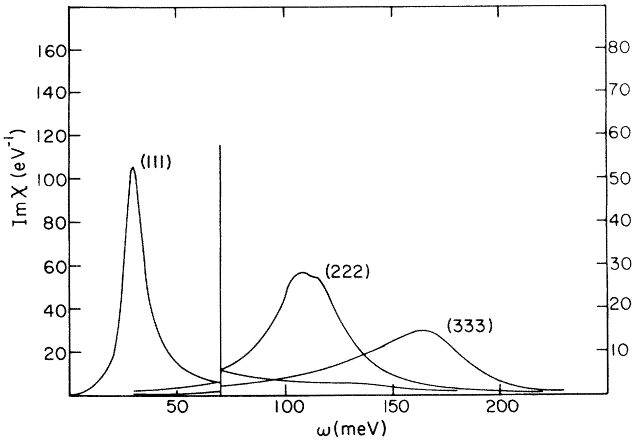

FIG. 3. Imaginary part of the transverse susceptibility for $\vec{p}$ along the [111] axis. The left energy scale refers to $\vec{p} = (2\pi/12a)(1,1,1)$; the right scale pertains to the two curves.

The calculations of Cooke, Lynn, and Davis[10] predicted the existence of an "optical" mode in the ⟨100⟩ direction which interacts with the "acoustical" mode in the neighborhood of $p = 0.25$ Å$^{-1}$, at an energy of about 150 meV. This mode was observed in the room-temperature measurements of Mook and Tochetti.[33] Our $T = 0$ calculations indicate a weak, broad optical spin-wave mode which for $p$ of about 0.25 Å$^{-1}$ has an energy of roughly 280 meV, about twice that reported in Ref. 33.

A second point of difference between our results and those of Ref. 10 is that although we observe some flattening of the spin-wave dispersion curve, we have not found $E(p)$ to decrease with increasing $p$ up to the highest values of $p$ considered here.

In addition, we find a small secondary maximum in the susceptibility for $\omega \approx 80$ meV in the case of $\vec{p} = (2\pi/a)(\frac{1}{12}, 0, 0)$. The real part of the determinant $D(p,\omega)$ has a local minimum in magnitude near this energy, but does not pass through zero. There is also a nearby maximum in the imaginary part of the non-self-consistent susceptibility. It is not clear whether this feature of the susceptibility would survive in a more refined calculation but its occurrence here indicates the possibility of struc-

TABLE II. Calculated energies and widths of spin waves.

| $\vec{p}$ $(\pi/6a)$ | $E$ (meV) | $\Gamma$ (meV) |
|---|---|---|
| (100) | 10.2 | 2.9 |
| (200) | 51.0 | 21.9 |
| (300) | 102 | 32.2 |
| (400) | 143 | 33.6 |
| (500) | 167 | 38.8 |
| (111) | 34.7 | 13.4 |
| (222) | 110.2 | 43.9 |
| (333) | 161.2 | 53.7 |

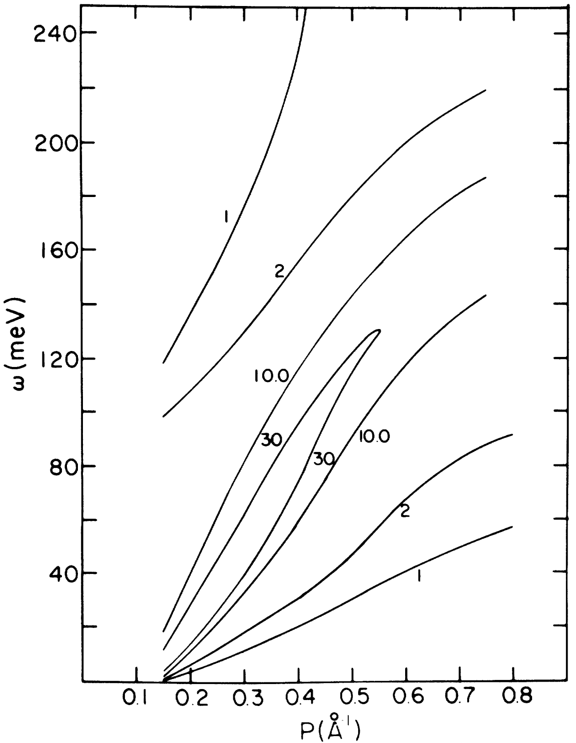

FIG. 4. Contour plot of the imaginary part of the transverse susceptibility for $\vec{p}$ along the [100] axis. The contours are labeled by the magnitude of $\mathrm{Im}(\chi^{+-}+\chi^{-+})$ in (eV atom)$^{-1}$.

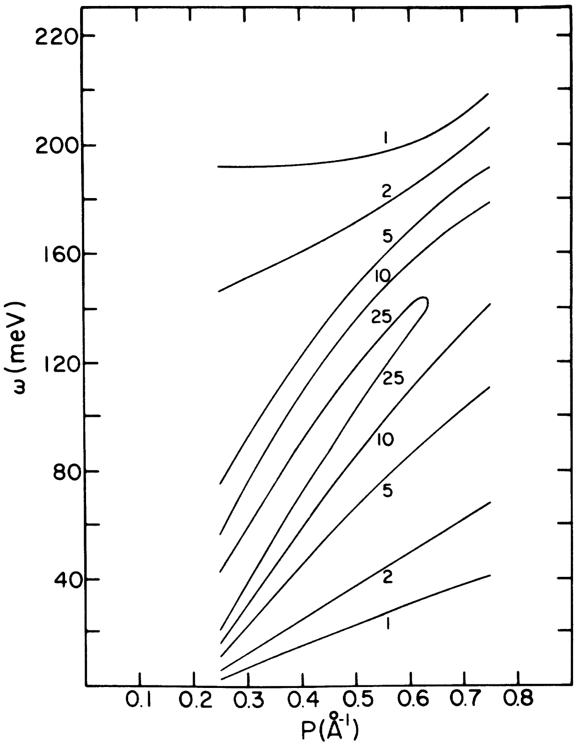

FIG. 5. Contour plot for $\vec{p}$ along the [111] axis.

ture in $\chi(p,\omega)$ which need not be associated clearly with spin waves.

Contour plots of the transverse susceptibility are shown in Figs. 4 and 5 for $\vec{p}$ along the [100] and [111] axis. These are necessarily fairly rough because only a small number of values of $p$ are available to us. However, the main features of this function appear, which includes gradual smearing of the spin-wave ridge. In comparison with the contours for momenta along the [100] axis, those for the [111] axis are broader, and have a lower maximum (less scattering intensity). This result appears to be in qualitative agreement with the experimental data, but comparison with the contours of scattering intensity at room temperature (Figs. 1 and 2 of Ref. 33) shows differences in detail: absence of optical modes, and a more gradual decrease in intensity (in the calculation) along the spin-wave ridge, particularly for $\vec{p}$ along [111]. It remains uncertain whether these differences result from temperature effects or indicate inadequacies in our parametrized calculation.

## VII. CONCLUSIONS

We have adapted the formal expression for magnetic susceptibilities previously derived from local spin-density-functional theory to enable use in connection with practical band calculations. This was done by transforming key parts of the calculation from a formulation based on an expansion in the reciprocal lattice to one involving an orbital basis.

Numerical evaluations have been made for the transverse spin susceptibility of nickel, based on a semiempirical band structure which is consistent with the experimental bands. Agreement with the general features (energies, widths, directional anisotropy) of the low-energy spin-wave spectrum is obtained by the choice of a single adjustable parameter. However, these are discrepancies in a detailed comparison with (mostly) room-temperature neutron scattering measurements, particularly in regard to optical modes, whose significance requires further investigation.

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