PHYSICAL REVIEW B VOLUME 15, NUMBER 11 1 JUNE 1977

Model calculation of dynamical spin susceptibility of paramagnetic nickel

Ramjit Singh and Satya Prakash*
Physics Department, Panjab University, Chandigarh, India
(Received 8 December 1975)

The dynamical spin susceptibility of paramagnetic nickel is investigated using a noninteracting isotropic band model based on the energy-band calculations of Hanus. The free-electron wave function and the simple tight-binding wave functions are used for s and d conduction electrons, respectively. Explicit expressions are obtained for the intraband and interband parts of the susceptibility function. Numerical computations are carried out for paramagnetic nickel for the atomic configuration $(3d)^{9.4}(4s)^{0.6}$ for field wave vector $\vec{q}$ along the [100] direction and energy-transfer range 0.01–0.4 eV. The intraband and interband parts are compared and the intraband part is found to be dominating. The exchange enhancement of the susceptibility function is also studied and the results are compared with other existing theoretical calculations and the experimental measurements. The agreement is found to be reasonably good.

I. INTRODUCTION

The quantum theory of the static susceptibility of simple metals has been put forward by many authors[1-5] through the use of pseudopotential formalism. Attention has recently been focused on the response of a metallic system to a spatially and time varying magnetic field, as this can directly be compared with neutron scattering measurements.[6] The formal functional forms of dynamical spin and orbital susceptibilities are rigorously obtained[7] but their explicit evaluation for a realistic multiband system is still awaited. Doniach[8] used the formalism due to Izuyama, Kim, and Kubo[9] to calculate the spin susceptibility of paramagnetic palladium in the random-phase approximation, in a one-band itinerant-electron model. Refinements of the model were subsequently considered by Allan *et al.*[10] and Lowde and Windsor[6] who calculated the spin susceptibility $\chi(\vec{q},\omega)$ of nickel in the paramagnetic and ferromagnetic phases. These authors used the free-electron and tight-binding approximations. In the free-electron approximation the role of overlap matrix elements in the expressions for the unenhanced susceptibility function is completely neglected. In the tight-binding approximation, the contribution of all the five d subbands is explicitly evaluated but the effects of an s-like band are completely ignored. Yamada and Shimizu[11] evaluated the dynamical spin susceptibility for a multiband system and calculated it, in the two-band model scheme for ferromagnetic nickel. Sokoloff[12] investigated the case of chromium in detail using a simplified free-electron-like band structure but Gupta and Sinha[13] calculated the static susceptibility of paramagnetic chromium using augmented-plane-wave band-structure calculations. Cooks and Wood[14] also calculated the static spin susceptibility function of ferromagnetic nickel using realistic band-structure calculations. Rath and Freeman[15] recently studied the static susceptibility of scandium using the augmented-plane-wave band-structure calculations. However, calculations are still awaited for the dynamical spin susceptibility of a transition metal.

It is a prohibitively difficult task to include the realistic multiband structure and actual crystal wave functions in the evaluation of the dynamical spin susceptibility $\chi(\vec{q},\omega)$ as this demands heavy computational efforts. Earlier the noninteracting-band model scheme was used successfully to investigate the dielectric screening, phonon frequencies, and effective ion-ion potential of noble and transition metals.[16-19] The model was also tested to investigate the wave-vector dependence of the spin susceptibility of paramagnetic nickel[20] and the results were found to be in good agreement with other detailed theoretical calculations. In view of the computational simplicity, we extend in this paper the formalism for the dynamical spin susceptibility and apply it to paramagnetic nickel. The plan of the paper is as follows: the formalism is presented in Sec. II. The calculations and the results are presented in Sec. III and these are discussed in Sec. IV.

II. THEORY

The frequency- and wave-vector-dependent spin susceptibility function in the random-phase approximation is given by the well-known expression

$$\chi^0(\vec{q}+\vec{G},\vec{q}+\vec{G}';\omega)=\frac{g^2\mu_B^2}{4}\sum_{\vec{k},\vec{k}'}\sum_{\substack{lm\sigma\\ l'm'\sigma'}}\frac{n_{lm\sigma}(\vec{k})-n_{l'm'\sigma'}(\vec{k}')}{E_{l'm'\sigma'}(\vec{k}')-E_{lm\sigma}(\vec{k})-\hbar\omega+i\epsilon}$$
$$\times\langle\Psi_{lm\sigma}(\vec{k})\,|\,e^{-i(\vec{q}+\vec{G})\cdot\vec{r}}\,|\,\Psi_{l'm'\sigma'}(\vec{k}')\rangle\langle\Psi_{l'm'\sigma'}(\vec{k}')\,|\,e^{i(\vec{q}+\vec{G}')\cdot\vec{r}}\,|\,\Psi_{lm\sigma}(\vec{k})\rangle\,, \tag{1}$$

where $n_{lm\sigma}(\vec{k})$ is the Fermi occupation probability function which is unity for an occupied state and zero otherwise. $\Psi_{lm\sigma}(\vec{k})$ and $E_{lm\sigma}(\vec{k})$ are the Bloch functions and energy eigenvalues for an electron with wave vector $\vec{k}$. l, m, and σ are the orbital, magnetic, and spin quantum numbers and act as band indices. $\vec{q}$ is the field wave vector and $\vec{G}$ and $\vec{G}'$ are the reciprocal-lattice vectors. ω is the frequency of the applied magnetic field, g is the Landé splitting factor, and μ_B is the Bohr magneton. ϵ is a small positive infinitesimal corresponding to an adiabatic turning on of the perturbing field. The summation $\vec{k}$ is over all the occupied electronic states. $\vec{k}$ and $\vec{k}'$ $(=\vec{k}+\vec{q})$ are restricted in the first Brillouin zone.

The susceptibility matrix consists of both the diagonal and nondiagonal parts, the latter giving rise to local-field corrections. It may be interesting to investigate both the diagonal and nondiagonal parts of the susceptibility matrix for a transition metal in which both the s and d electrons are itinerant. Because there exist other theoretical calculations only for the diagonal part, therefore we limit our calculations to the evaluation of the diagonal part of the susceptibility matrix. Retaining only the normal-process contributions Eq. (1) reduces to

$$\chi^0(\vec{q},\omega)=\frac{g^2\mu_B^2}{2}\sum_{\vec{k},\vec{k}'}\sum_{\substack{lm\\ l'm'}}\frac{n_{lm}(\vec{k})-n_{l'm'}(\vec{k}')}{E_{l'm'}(\vec{k}')-E_{lm}(\vec{k})-\hbar\omega+i\epsilon}$$
$$\times\left|\langle\Psi_{lm}(\vec{k})\,|\,e^{-i\vec{q}\cdot\vec{r}}\,|\,\Psi_{l'm'}(\vec{k}')\rangle\right|^2. \tag{2}$$

In Eq. (2), the orthonormality of spin wave functions is used. The spin index σ has been dropped for a paramagnetic system and the right-hand side of Eq. (2) is multiplied by a factor of 2 for spin degeneracy.

In a transition metal the itinerant electrons are distributed in the s and d subbands. These electrons undergo the intra- and interband transitions and readjust themselves in response to the applied magnetic field. We can, therefore write the susceptibility function as

$$\chi^0(\vec{q},\omega)=\chi^0_{ss}(\vec{q},\omega)+\chi^0_{dd}(\vec{q},\omega)+\chi^0_{ds}(\vec{q},\omega)+\chi^0_{sd}(\vec{q},\omega)\,, \tag{3}$$

where the expressions for $\chi^0_{ss}(\vec{q},\omega)$, etc., can readily be written with the help of Eq. (2). $\chi^0_{ss}(\vec{q},\omega)$, $\chi^0_{dd}(\vec{q},\omega)$, $\chi^0_{ds}(\vec{q},\omega)$, and $\chi^0_{sd}(\vec{q},\omega)$ correspond to the transitions from s band to s band, from d subbands to d subbands, from d subbands to s band, and from s band to d subbands, respectively.

A. Evaluation of $\chi^0_{ss}(\vec{q},\omega)$

We use the free-electron approximation for the wave function and the parabolic-band approximation for the energy eigenvalues, for the electrons in the s band. The real and the imaginary parts of the spin susceptibility function are separated using the identity

$$\lim_{\epsilon\to 0}\,(x\pm i\epsilon)^{-1}\equiv 1/x\mp i\pi\delta(x)\,, \tag{4}$$

and the sum over $\vec{k}$ is replaced by integration. We get the explicit expressions for the real and the imaginary parts as follows:

$$\mathrm{Re}\chi^0_{ss}(\vec{q},\omega)=\frac{g^2\mu_B^2N\Omega_0}{4\pi^2}m_sk_{Fs}\left\{\frac{1}{2}+\frac{1}{8\lambda}\left[1-\left(\frac{m_sw}{\lambda}-\lambda\right)^2\right]\ln\left|\frac{1-m_sw/\lambda+\lambda}{1+m_sw/\lambda-\lambda}\right|-\frac{1}{8\lambda}\left[1-\left(\frac{m_sw}{\lambda}+\lambda\right)^2\right]\ln\left|\frac{1-m_sw/\lambda-\lambda}{1+m_sw/\lambda+\lambda}\right|\right\}, \tag{5}$$

$$\mathrm{Im}\chi^0_{ss}(\vec{q},\omega)=-\frac{g^2\mu_B^2N\Omega_0}{32\pi\lambda}m_sk_{Fs}\left\{\left[1-\left(\frac{m_sw}{\lambda}-\lambda\right)^2\right]\theta\left(1-\left(\frac{m_sw}{\lambda}-\lambda\right)^2\right)-\left[1-\left(\frac{m_sw}{\lambda}+\lambda\right)^2\right]\theta\left(1-\left(\frac{m_sw}{\lambda}+\lambda\right)^2\right)\right\}, \tag{6}$$

where

$$\lambda=q/2k_{Fs}\,,\quad w=\omega/2k_{Fs}^2 \tag{7}$$

and $\theta(x)$ is step function which is unity if $x\geq 0$ and zero otherwise. N is the number of unit cells in the crystal and Ω_0 is the volume of the unit cell. m_s and k_{Fs} are the effective mass and Fermi momentum of s electrons. Equations (5) and (6) are similar to those obtained by Doniach.[8]

B. Evaluation of $\chi^0_{dd}(\vec{q},\omega)$

Using the simple tight-binding wave function and parabolic band approximation for the itinerant d-electrons,[16,21] we get

$$\chi^0_{dd}(\vec{q},\omega) = \frac{g^2\mu_B^2}{2}\sum_{\vec{k}}\sum_{m,m'}\left(\frac{n_{dm}(\vec{k})-n_{dm'}(\vec{k}+\vec{q})}{E_{dm'}(\vec{k}+\vec{q})-E_{dm}(\vec{k})-\hbar\omega+i\epsilon}\right)\times[\Delta_{dm,dm'}(\vec{q})\,\Delta^*_{dm,dm'}(\vec{q})]\,, \tag{8}$$

where the explicit expressions for the overlap matrix elements $\Delta_{dm,dm'}(\vec{q})$ are the same as given by Prakash and Joshi.[21] Here the overlapping between d wave functions on the same atomic sites is included and the overlapping on the different atomic sites is neglected. Therefore $\Delta_{dm,dm'}(\vec{q})$ reduces exactly to what we call the atomic form factors. Only the normal-process contribution is retained[22] in the derivation of Eq. (8). We call the first factor in parentheses in Eq. (8), the band-structure part, and the second factor in square brackets, the overlap-integral part.

For intra-d-subband transitions, i.e., when $m=m'$, we evaluate $\chi^0_{dd}(\vec{q},\omega)$ exactly in the same manner as $\chi^0_{ss}(\vec{q},\omega)$. The expressions for the real and imaginary parts are

$$\mathrm{Re}\chi^0_{dd(\mathrm{intra})}(\vec{q},\omega) = \frac{g^2\mu_B^2 N\Omega_0}{4\pi^2}\sum_m m_{dm}k_{Fdm}I_{dm}\left\{\frac{1}{2}+\frac{1}{8\lambda'}\left[1-\left(\frac{m_{dm}w'}{\lambda'}-\lambda'\right)^2\right]\ln\left|\frac{1-m_{dm}w'/\lambda'+\lambda'}{1+m_{dm}w'/\lambda'-\lambda'}\right|\right.$$
$$\left.-\frac{1}{8\lambda'}\left[1-\left(\frac{m_{dm}w'}{\lambda'}+\lambda'\right)^2\right]\ln\left|\frac{1-m_{dm}w'/\lambda'-\lambda'}{1+m_{dm}w'/\lambda'+\lambda'}\right|\right\}, \tag{9}$$

$$\mathrm{Im}\chi^0_{dd(\mathrm{intra})}(\vec{q},\omega) = -\frac{g^2\mu_B^2 N\Omega_0}{32\pi}\sum_m\frac{m_{dm}k_{Fdm}}{\lambda'}I_{dm}\left\{\left[1-\left(\frac{m_{dm}w'}{\lambda'}-\lambda'\right)^2\right]\theta\left(1-\left(\frac{m_{dm}w'}{\lambda'}-\lambda'\right)^2\right)\right.$$
$$\left.-\left[1-\left(\frac{m_{dm}w'}{\lambda'}+\lambda'\right)^2\right]\theta\left(1-\left(\frac{m_{dm}w'}{\lambda'}+\lambda'\right)^2\right)\right\}, \tag{10}$$

where

$$\lambda' = q/2k_{Fdm}\,,\quad w' = \omega/2k^2_{Fdm}\,,\quad I_{dm} = \Delta_{dm,dm'}(\vec{q})\Delta^*_{dm,dm'}(\vec{q})\,. \tag{11}$$

m_{dm} and k_{Fdm} are the effective mass and Fermi momentum for the d subband with magnetic quantum number m and $\theta(x)$ is the step function as defined earlier. In the $\vec{q}\to 0$ and $\omega\to 0$ limits, the intraband part reduces to the value of the density of states of d electrons at the Fermi surface. It was pointed out by Gupta and Sinha[13] and by Prakash and Singh[20] that the overlap matrix elements reduce the magnitude of susceptibility and also change the nature of susceptibility function.

For the inter-d-subband transitions, i.e., when $m\neq m'$, the analytical expressions for real and imaginary parts of the susceptibility function are as follows:

$$\chi^0_{dd(\mathrm{inter})}(\vec{q},\omega) = \sum_{m\neq m'}(I_1+iI_2)\Delta_{dm,dm'}(\vec{q})\,\Delta^*_{dm,dm'}(\vec{q})\,, \tag{12}$$

where

$$I_1 = -\frac{g^2\mu_B^2 N\Omega_0}{4\pi^2}\left[m_{dm'}\left(\frac{k_{Fdm}}{\xi}-X_1-\frac{1}{4\xi^2}Y_1\right)+m_{dm}\left(\frac{k_{Fdm'}}{\xi'}-X_2-\frac{1}{4\xi'^2}Y_2\right)\right]\,, \tag{13}$$

$$X_1 = \left[\frac{k^2_{Fdm}}{4q}-\frac{q}{4\xi}\left(1+\frac{2}{\xi}\right)+\frac{m_{dm'}\omega}{2\xi q}\right]\ln\left|\frac{2k_{Fdm}q-\xi k^2_{Fdm}+q^2-2m_{dm'}\omega}{2k_{Fdm}q+\xi k^2_{Fdm}-q^2+2m_{dm'}\omega}\right|\,, \tag{14}$$

$$X_2 = \left[\frac{k^2_{Fdm'}}{4q}-\frac{q}{4\xi'}\left(1+\frac{2}{\xi'}\right)-\frac{m_{dm}\omega}{2\xi' q}\right]\ln\left|\frac{2k_{Fdm'}q-\xi' k^2_{Fdm'}+q^2+2m_{dm}\omega}{2k_{Fdm'}q+\xi' k^2_{Fdm'}-q^2-2m_{dm}\omega}\right|\,, \tag{15}$$

$$Y_1 = \Delta\times\begin{cases}\dfrac{2}{\sqrt{\Delta}}\left(\tan^{-1}\dfrac{2(\xi k_{Fdm}+q)}{\sqrt{\Delta}}+\tan^{-1}\dfrac{2(\xi k_{Fdm}-q)}{\sqrt{\Delta}}\right) & \text{if } \Delta>0\\[2ex] \dfrac{1}{\sqrt{-\Delta}}\left(\ln\left|\dfrac{2\xi k_{Fdm}+2q-\sqrt{-\Delta}}{2\xi k_{Fdm}+2q+\sqrt{-\Delta}}\right|+\ln\left|\dfrac{2\xi k_{Fdm}-2q-\sqrt{-\Delta}}{2\xi k_{Fdm}-2q+\sqrt{-\Delta}}\right|\right) & \text{if } \Delta<0\,,\end{cases} \tag{16}$$

$$Y_2=\Delta'\times\begin{cases}\dfrac{2}{(\Delta')^{1/2}}\left(\tan^{-1}\dfrac{2(\xi' k_{Fdm'}+q)}{(\Delta')^{1/2}}+\tan^{-1}\dfrac{2(\xi' k_{Fdm'}-q)}{(\Delta')^{1/2}}\right) & \text{if } \Delta'>0\\ (\Delta')^{-1/2}\left(\ln\left|\dfrac{2\xi' k_{Fdm'}+2q-(-\Delta')^{1/2}}{2\xi' k_{Fdm'}+2q+(-\Delta')^{1/2}}\right|+\ln\left|\dfrac{2\xi' k_{Fdm'}-2q-(-\Delta')^{1/2}}{2\xi' k_{Fdm'}-2q+(-\Delta')^{1/2}}\right|\right) & \text{if } \Delta'<0\,,\end{cases} \tag{17}$$

$$\Delta=-4[q^2(\xi+1)-2m_{dm'}\omega\xi]\,, \tag{18}$$

$$\Delta'=-4[q^2(\xi'+1)+2m_{dm}\omega\xi']\,, \tag{19}$$

$$\xi=m_{dm'}/m_{dm}-1;\quad \xi'=m_{dm}/m_{dm'}-1\,,$$

and

$$I_2=-\frac{g^2\mu_B^2 N\Omega_0}{16\pi q}(Z_1+Z_2)\,, \tag{20}$$

$$Z_1=m_{dm'}\times\begin{cases}k_{Fdm}^2-k_1^2 & \text{if } k_1<k_{Fdm}<k_2;\quad \mu_1\geq 0\\ k_2^2-k_1^2 & \text{if } k_2<k_{Fdm};\quad \mu_1\geq 0\\ 0 & \text{if } k_{Fdm}<k_1;\quad \mu_1\geq 0\\ 0 & \text{if } \mu_1<0;\end{cases} \tag{21}$$

$$Z_2=m_{dm}\times\begin{cases}k_{Fdm'}^2-k_1'^2 & \text{if } k_1'<k_{Fdm'}<k_2';\quad \mu_2\geq 0\\ k_2'^2-k_1'^2 & \text{if } k_2'<k_{Fdm'};\quad \mu_2\geq 0\\ 0 & \text{if } k_{Fdm'}<k_1';\quad \mu_2\geq 0\\ 0 & \text{if } \mu_2<0;\end{cases} \tag{22}$$

$$\mu_1=-\Delta/4m_{dm'}^2\,, \tag{23}$$

$$\mu_2=-\Delta'/4m_{dm}^2\,, \tag{24}$$

$$k_1=q/\xi-\sqrt{-\Delta}/2\xi\,, \tag{25}$$

$$k_2=q/\xi+\sqrt{-\Delta}/2\xi\,, \tag{26}$$

$$k_1'=q/\xi'-(-\Delta')^{1/2}/2\xi'\,, \tag{27}$$

and

$$k_2'=q/\xi'+(-\Delta')^{1/2}/2\xi'\,. \tag{28}$$

These expressions are similar to those obtained by Czachor[23] for the interband transitions in the evaluation of the dielectric function for a semimetal. In the limit $q\to 0$, $\chi^0_{dd(\text{inter})}(\vec{q},\omega)$ reduces to zero as $\Delta_{dm,dm'}(0)=0$, because of the orthogonality of the d wave functions.

Izuyama, Kim, and Kubo[9] pointed out that the reduced susceptibility can be related to the neutron scattering measurements, therefore for comparison we also calculate the reduced spin susceptibility. Our results for intra- and inter-d-subband transitions directly reduce to the reduced spin susceptibility if we take the atomic factor $\Delta_{dm,dm'}(\vec{q})$ to be unity. These expressions become equivalent in principle to those obtained by Allan *et al.*[10] and Lowde and Windsor.[6]

C. Evaluation of $\chi^0_{ds}(\vec{q},\omega)$

Using the free-electron approximation for s electrons and the simple tight-binding wave function for d electrons, we get

$$\chi^0_{ds}(\vec{q},\omega)=\frac{g^2\mu_B^2}{2}\frac{1}{\Omega_0}\sum_{\vec{k}}\sum_m\frac{n_{dm}(\vec{k})-n_s(\vec{k}+\vec{q})}{E_s(\vec{k}+\vec{q})-E_{dm}(\vec{k})-\hbar\omega+i\epsilon}\times\int\Phi^*_{dm}(\vec{r})\,e^{i\vec{k}\cdot\vec{r}}\,d\vec{r}\times\int\Phi_{dm}(\vec{r})e^{-i\vec{k}\cdot\vec{r}}\,d\vec{r}. \tag{29}$$

The real and imaginary parts are separated using the identity (4) and are evaluated exactly in the same manner as done by Prakash and Joshi[21] and by Prakash and Singh[20] for the static susceptibility. The explicit expressions for real and imaginary parts of $\chi^0_{ds}(\vec{q},\omega)$ are as follows:

$$\mathrm{Re}\chi^0_{ds}(\vec{q},\omega)=\frac{2g^2\mu_B^2 N m_s(48)^2}{\pi}\times\sum_i\sum_j a_i a_j\alpha_i\alpha_j\sum_m(-1)^m\int_0^{k_{Fdm}}\frac{dk\,k^6}{(k^2+\alpha_i^2)^4(k^2+\alpha_j^2)^4}\times[D^2_{0m}D^2_{0-m}I^1_0+(D^2_{1m}D^2_{-1-m}+D^2_{-1m}D^2_{1-m})I^1_1+(D^2_{2m}D^2_{-2-m}+D^2_{-2m}D^2_{2-m})I^1_2]\,, \tag{30}$$

$$\mathrm{Im}\chi^0_{ds}(\vec{q},\omega)=-2g^2\mu_B^2 N m_s(48)^2\times\sum_i\sum_j a_i a_j\alpha_i\alpha_j\sum_m(-1)^m\int_0^{k_{Fdm}}\frac{dk\,k^6}{(k^2+\alpha_i^2)^4(k^2+\alpha_j^2)^4}\times[D^2_{0m}D^2_{0-m}I^2_0+(D^2_{1m}D^2_{-1-m}+D^2_{-1m}D^2_{1-m})I^2_1+(D^2_{2m}D^2_{-2-m}+D^2_{-2m}D^2_{2-m})I^2_2]\,. \tag{31}$$

Here $D^2_{m'm}$ are the elements of the rotation matrix with argument $(-\gamma, -\beta, -\alpha)$ while α, β, and γ are Euler's angles. a_i and α_i are the parameters of the $3d$ radial wave function.[24]

$$I^1_0 = \tfrac{5}{4}(\tfrac{1}{2}I_{n0} - 3I_{n2} + \tfrac{9}{2}I_{n4}), \tag{32a}$$

$$I^1_1 = \tfrac{15}{4}(-I_{n2} + I_{n4}), \tag{32b}$$

$$I^1_2 = \tfrac{15}{8}(\tfrac{1}{2}I_{n0} - I_{n2} + \tfrac{1}{2}I_{n4}), \tag{32c}$$

$$I_{n0} = -\frac{1}{b}\ln\left|\frac{b-a}{b+a}\right|, \tag{33a}$$

$$I_{n2} = -\frac{1}{b}\left(\frac{2a}{b} + \frac{a^2}{b^2}\ln\left|\frac{b-a}{b+a}\right|\right), \tag{33b}$$

$$I_{n4} = -\frac{1}{b}\left(\frac{2a}{3b} + \frac{2a^3}{b^3} + \frac{a^4}{b^4}\ln\left|\frac{b-a}{b+a}\right|\right), \tag{33c}$$

$$I^2_0 = \begin{cases} \dfrac{5}{8b}\left(\dfrac{3a^2}{b^2} - 1\right)^2 & \text{if} \left(1 - \dfrac{a^2}{b^2}\right) \geq 0, \\ 0 & \text{if} (1 - a^2/b^2) < 0; \end{cases} \tag{34a}$$

$$I^2_1 = \begin{cases} \dfrac{15}{4b}\dfrac{a^2}{b^2}\left(\dfrac{a^2}{b^2} - 1\right) & \text{if} \left(1 - \dfrac{a^2}{b^2}\right) \geq 0, \\ 0 & \text{if} (1 - a^2/b^2) < 0; \end{cases} \tag{34b}$$

$$I^2_2 = \begin{cases} \dfrac{15}{16b}\left(\dfrac{a^2}{b^2} - 1\right)^2 & \text{if} \left(1 - \dfrac{a^2}{b^2}\right) \geq 0, \\ 0 & \text{if} (1 - a^2/b^2) < 0; \end{cases} \tag{34c}$$

$$a = (1 - m_s/m_{dm})k^2 + q^2 - 2m_s\omega, \tag{35}$$

and

$$b = 2kq. \tag{36}$$

The radial integration over $\vec{k}$ in Eqs. (30) and (31) is carried out numerically. The rotation matrices and the parameters of the $3d$ radial wave function are taken from the paper of Prakash and Joshi.[21]

$\chi^0_{ds}(\vec{q}, \omega)$ should reduce to zero in the $\vec{q} \to 0$ limit if an orthogonal set of eigenfunctions is used for s and d conduction electrons. However in our calculations, the s and d wave functions are not orthogonal, therefore $\chi^0_{ds}(\vec{q}, \omega)$ does not reduce exactly to zero at $\vec{q} = 0$ but we have taken it to be zero at $\vec{q} = 0$ and evaluated it exactly for finite values of $\vec{q}$.

Because of the $\vec{k}$ dependence of the overlap integrals, it is not possible to separate the band-structure and overlap-integral parts of $\chi^0_{ds}(\vec{q}, \omega)$. Therefore it is not straightforward to have an expression for the reduced susceptibility as we did in the case of $\chi^0_{dd}(\vec{q}, \omega)$. However, to have an expression for the reduced susceptibility which may be compared with the experimental data, we take $k = k_{Fdm}$ in the overlap integrals and take them out of the integral over k and put that equal to unity to obtain $\bar{\chi}^0_{ds}(\vec{q}, \omega)$. This approximation is justified because the maximum response of electrons is due to the electrons in the vicinity of the Fermi surface. However, this may underestimate the reduced $\bar{\chi}^0_{ds}(\vec{q}, \omega)$, yet the final results may not be seriously affected because the contribution of $\chi^0_{ds}(\vec{q}, \omega)$ is itself very small as found in the calculation of the static susceptibility[20] and dielectric function of nickel.[21] The expressions for the real and imaginary parts of the reduced susceptibility thus obtained, are as follows:

$$\mathrm{Re}\bar{\chi}^0_{ds}(\vec{q}, \omega) = -\frac{g^2\mu_B^2 N\Omega_0 M_s}{4\pi^2}\sum_m\left(\frac{k_{Fdm}}{\xi''} - X_3 - \frac{1}{4\xi''^2}Y_3\right), \tag{37}$$

$$\mathrm{Im}\bar{\chi}^0_{ds}(\vec{q}, \omega) = -\frac{g^2\mu_B^2 N\Omega_0 M_s}{16\pi q} \times \sum_m \begin{cases} k^2_{Fdm} - k''^2_1 & \text{if } k''_1 < k_{Fdm} < k''_2; \quad \mu' \geq 0 \\ k''^2_2 - k''^2_1 & \text{if } k''_2 < k_{Fdm}; \quad \mu' \geq 0 \\ 0 & \text{if } k_{Fdm} < k''_1; \quad \mu' \geq 0 \\ 0 & \text{if } \mu' < 0 \end{cases} \tag{38}$$

where

$$X_3 = \left[\frac{k^2_{Fdm}}{4q} - \frac{q}{4\xi''}\left(1 + \frac{2}{\xi''}\right) + \frac{m_s\omega}{2\xi'' q}\right] \times \ln\left|\frac{2k_{Fdm}q - \xi''k^2_{Fdm} + q^2 - 2m_s\omega}{2k_{Fdm}q + \xi''k^2_{Fdm} - q^2 + 2m_s\omega}\right|, \tag{39}$$

$$Y_3 = \Delta'' \times \begin{cases} \dfrac{2}{(\Delta'')^{1/2}}\left(\tan^{-1}\dfrac{2(\xi''k_{Fdm} + q)}{(\Delta'')^{1/2}} + \tan^{-1}\dfrac{2(\xi''k_{Fdm} - q)}{(\Delta'')^{1/2}}\right) & \text{if } \Delta'' > 0, \\ \dfrac{1}{(-\Delta'')^{1/2}}\left(\ln\left|\dfrac{2\xi''k_{Fdm} + 2q - (-\Delta'')^{1/2}}{2\xi''k_{Fdm} + 2q + (-\Delta'')^{1/2}}\right| + \ln\left|\dfrac{2\xi''k_{Fdm} - 2q - (-\Delta'')^{1/2}}{2\xi''k_{Fdm} - 2q + (-\Delta'')^{1/2}}\right|\right) & \text{if } \Delta'' < 0, \end{cases} \tag{40}$$

$$\mu' = (m_s^2)^{-1}[q^2(\xi'' + 1) - 2m_s\omega\xi''], \tag{41}$$

$$\Delta'' = -4\mu', \tag{42}$$

$$k''_1 = q/\xi'' - (\mu')^{1/2}/\xi'', \tag{43}$$

$$k''_2 = q/\xi'' + (\mu')^{1/2}/\xi'', \tag{44}$$

and

$$\xi'' = m_s/m_{dm} - 1. \tag{45}$$

D. Evaluation of $\chi^0_{sd}(\vec{q},\omega)$

The expressions for the real and the imaginary parts of $\chi^0_{sd}(\vec{q},\omega)$ are evaluated exactly in the same manner as for $\chi^0_{ds}(\vec{q},\omega)$. In this case the transitions to be considered are from s band to d subbands. The explicit expressions for the real and the imaginary parts are as follows:

$$\mathrm{Re}\chi^0_{sd}(\vec{q},\omega)=\frac{2g^2\mu_B^2N(48)^2}{\pi}\sum_i\sum_j a_ia_j\alpha_i\alpha_j\sum_m(-1)^m m_{dm}\int_0^{k_{Fs}}dk\,k^2[D^2_{0m}D^2_{0-m}I^3_0+(D^2_{1m}D^2_{-1-m}+D^2_{-1m}D^2_{1-m})I^3_1 + (D^2_{2m}D^2_{-2-m}+D^2_{-2m}D^2_{2-m})I^3_2], \tag{46}$$

$$\mathrm{Im}\chi^0_{sd}(\vec{q},\omega)=-2\,g^2\mu_B^2N(48)^2\sum_i\sum_j a_ia_j\alpha_i\alpha_j\sum_m(-1)^m m_{dm}\int_0^{k_{Fs}}dk\,k^2[D^2_{0m}D^2_{0-m}I^4_0+(D^2_{1m}D^2_{-1-m}+D^2_{-1m}D^2_{1-m})I^4_1 + (D^2_{2m}D^2_{-2-m}+D^2_{-2m}D^2_{2-m})I^4_2]. \tag{47}$$

Here

$$I^3_0=\frac{5}{8}\int_{-1}^{1}\frac{(3k^2t^2+2bt+2q^2-k^2)^2}{S'(a'+bt)}\,dt, \tag{48}$$

$$I^3_1=\frac{15}{4}\int_{-1}^{1}\frac{k^2(t^2-1)(kt+q)^2}{S'(a'+bt)}\,dt, \tag{49}$$

$$I^3_2=\frac{15}{16}\int_{-1}^{1}\frac{k^4(t^2-1)^2}{S'(a'+bt)}\,dt, \tag{50}$$

$$I^4_0=\begin{cases}\dfrac{5}{8b}\left[3\left(\dfrac{a'^2}{b^2}-\dfrac{1}{3}\right)k^2+2q^2-2a'\right]^2\Big/S & \text{if } \left(1-\dfrac{a'^2}{b^2}\right)\geq 0\\ 0 & \text{if } (1-a'^2/b^2)<0,\end{cases} \tag{51}$$

$$I^4_1=\begin{cases}\dfrac{15}{4b}\left[k^2\left(\dfrac{a'^2}{b^2}-1\right)\left(\dfrac{k^2a'^2}{b^2}+q^2-a'\right)\right]\Big/S & \text{if } \left(1-\dfrac{a'^2}{b^2}\right)\geq 0\\ 0 & \text{if } (1-a'^2/b^2)<0,\end{cases} \tag{52}$$

$$I^4_2=\begin{cases}\dfrac{15}{16b}\left[k^4\left(\dfrac{a'^2}{b^2}-1\right)^2\right]\Big/S & \text{if } \left(1-\dfrac{a'^2}{b^2}\right)\geq 0\\ 0 & \text{if } (1-a'^2/b^2)<0,\end{cases} \tag{53}$$

$$a'=(1-m_{dm}/m_s)k^2+q^2-2m_{dm}\omega, \tag{54}$$

$$S=(k^2+q^2+\alpha_i^2-a')^4(k^2+q^2+\alpha_j^2-a')^4, \tag{55}$$

$$S'=(k^2+q^2+\alpha_i^2+bt)^4(k^2+q^2+\alpha_j^2+bt)^4.$$

The integrations in the expressions for I^3_0, I^3_1, and I^3_2 are carried out using the method of partial fractions. The expressions for these partial fractions are very lengthy and therefore these are not given in the text. $\chi^0_{sd}(\vec{q},\omega)$ also does not reduce to correct limit for $\vec{q}\to 0$, but expressions are exact for finite values of $\vec{q}$.

The expressions for the real and imaginary parts of the reduced susceptibility are also obtained in the same manner as for $\bar{\chi}^0_{ds}(\vec{q},\omega)$ and these are given as follows:

$$\mathrm{Re}\bar{\chi}^0_{sd}(\vec{q},\omega)=-\frac{g^2\mu_B^2N\Omega_0}{4\pi^2}\times\sum_m m_{dm}\left(\frac{k_{Fdm}}{\xi'''}-X_4-\frac{4}{\xi'''^2}Y_4\right), \tag{56}$$

$$\mathrm{Im}\bar{\chi}^0_{sd}(\vec{q},\omega)=-\frac{g^2\mu_B^2N\Omega_0}{16\pi q}\times\sum_m m_{dm}\times\begin{cases}k^2_{Fs}-k'''^2_1 & \text{if } k'''_1<k_{Fs}<k'''_2;\ \mu''\geq 0\\ k'''^2_2-k'''^2_1 & \text{if } k'''_2<k_{Fs};\ \mu''\geq 0\\ 0 & \text{if } k_{Fs}<k'''_1;\ \mu''\geq 0,\\ 0 & \text{if } \mu''\leq 0;\end{cases} \tag{57}$$

where

$$X_4=\left[\frac{k_{Fs}^2}{4q}-\frac{q}{4\xi'''}\left(1+\frac{2}{\xi'''}\right)+\frac{m_{dm}\omega}{2\xi'''q}\right]\times\ln\left|\frac{2k_{Fs}q-\xi'''k^2_{Fs}+q^2-2m_{dm}\omega}{2k_{Fs}q+\xi'''k^2_{Fs}-q^2+2m_{dm}\omega}\right|, \tag{58}$$

$$Y_4=\Delta'''\times\begin{cases}\dfrac{2}{(\Delta''')^{1/2}}\left(\arctan\dfrac{2(\xi'''k_{Fs}+q)}{(\Delta''')^{1/2}}+\arctan\dfrac{2(\xi'''k_{Fs}-q)}{(\Delta''')^{1/2}}\right) & \text{if } \Delta'''>0\\ (-\Delta''')^{-1/2}\left(\ln\left|\dfrac{2\xi'''k_{Fs}+2q-(-\Delta''')^{1/2}}{2\xi'''k_{Fs}+2q+(-\Delta''')^{1/2}}\right|+\ln\left|\dfrac{2\xi'''k_{Fs}-2q-(-\Delta''')^{1/2}}{2\xi'''k_{Fs}-2q+(-\Delta''')^{1/2}}\right|\right) & \text{if } \Delta'''<0,\end{cases} \tag{59}$$

$$\mu''=(m^2_{dm})^{-1}[q^2(\xi'''+1)-2m_{dm}\omega\xi'''], \tag{60}$$

$$\Delta'''=-4\mu'', \tag{61}$$

$$k_1''' = q/\xi''' - (\mu''')^{1/2}/\xi''', \tag{62}$$

$$k_2''' = q/\xi''' + (\mu''')^{1/2}/\xi''', \tag{63}$$

and

$$\xi''' = m_{dm}/m_s - 1.$$

III. CALCULATIONS AND RESULTS

A. Model band structure

We use the noninteracting-band model due to Prakash and Joshi[21] and use the same values for physical parameters, effective masses, and Fermi momenta. Prakash and Joshi assign a fixed magnetic quantum number m to a particular d subband. But in the construction of the isotropic-d-band model, the d components of the wave function hybridize and this m assignment does not remain valid. Therefore we count the bands from one to five in the ascending order of energy and give equal weight to every m component of the wave function for each d subband while evaluating the susceptibility function. Such an averaging was also used in the earlier calculations of the dielectric function for nickel, chromium, vanadium, platinum, and palladium.[25]

B. Unenhanced susceptibility function

In the present calculation we fix $\vec{q}$ along the [100] direction and evaluate the real and imaginary parts of the susceptibility function of paramagnetic nickel for the atomic configuration $(3d)^{9.4}(4s)^{0.6}$. The results for the different values of $\vec{q}$ in the energy-transfer range 0.01 to 0.4 eV are shown in Fig. 1, where the inter- and the intraband parts are also shown separately by dashed lines. The contribution of the interband part varies from 1 to 10% that of the intraband part for different momentum transfers. The interband part also shows oscillatory nature. For $\vec{q}=(\frac{1}{18})$ [100], $\mathrm{Re}\chi^0(\vec{q},\omega)$ decreases rapidly and becomes negative for $\hbar\omega \geq 0.23$ eV. Its magnitude again starts increasing beyond $\hbar\omega > 0.28$ eV. The imaginary part increases almost linearly and shows a singular behavior in the vicinity of $\hbar\omega = 0.26$ eV. This anomalous behavior is found only in the intraband part. The interband part does not show such anomalies. As the value of $\vec{q}$ increases, the anomalous behavior starts vanishing. The intraband part of $\mathrm{Re}\chi^0(\vec{q},\omega)$ becomes almost constant but the interband part still shows the oscillatory nature which is responsible for the broad features in the susceptibility function for $\hbar\omega = 0.12$, 0.21, and 0.31 eV when $\vec{q}=(\frac{7}{18})$ [100] and for $\hbar\omega = 0.12$, 0.27, and 0.36 eV when $\vec{q}=$[100]. For higher values of $\vec{q}$, $\mathrm{Im}\chi^0(\vec{q},\omega)$ increases with increasing values of ω. The oscillatory behavior is absent in both the intra- and interband parts. The qualitative behavior presented by our results is similar to that obtained by Julien *et al.*[26] and by Hebborn and March.[7]

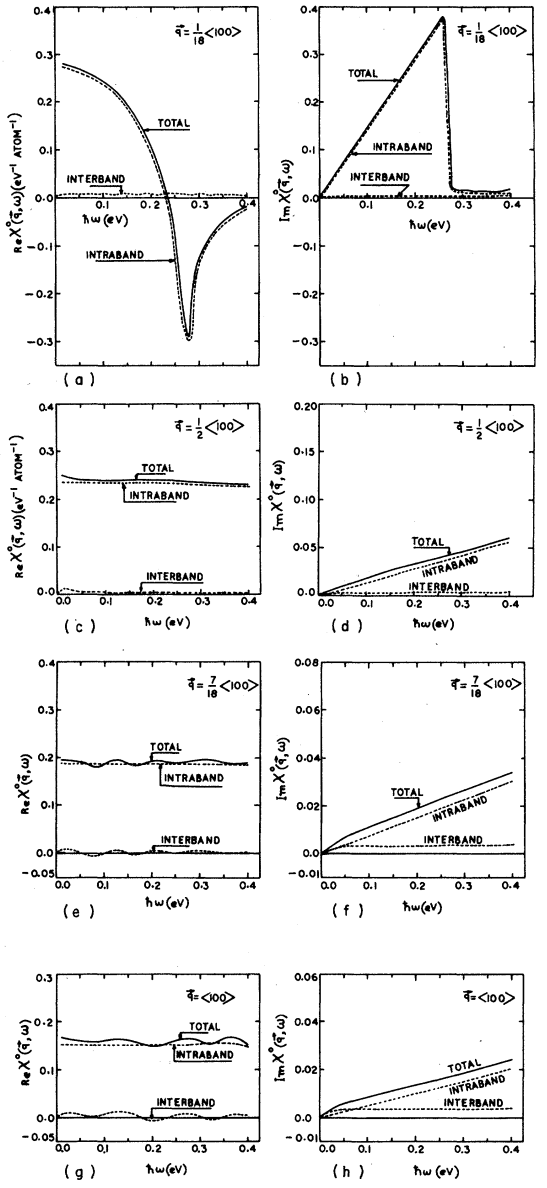

FIG. 1. $\chi^0(\vec{q},\omega)$ vs $\hbar\omega$ for paramagnetic nickel. Dashed lines represent the intra- and interband parts and the solid line represents the total susceptibility. $\chi^0(\vec{q},\omega)$ is measured in units of $g^2\mu_B{}^2\,\mathrm{eV}^{-1}$ per atom.

The real and imaginary parts of the susceptibility function are also studied as a function of $\vec{q}$ for different values of energy transfer. These results are shown in Fig. 2. For low-energy transfers $\mathrm{Re}\chi^0(\vec{q},\omega)$ decreases with increasing $\vec{q}$ but for higher-energy transfers, the susceptibility function shows a broad maximum at $\vec{q}=0.47$. The

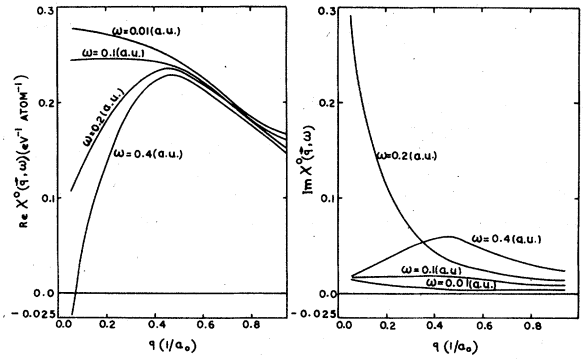

FIG. 2. $\chi^0(\vec{q},\omega)$ vs $\vec{q}$ along [100] for paramagnetic nickel. $\vec{q}$ is measured in inverse Bohr units.

imaginary part of the susceptibility function also decreases for increasing values of $\vec{q}$ for low-energy transfers and shows a broad maximum for high-energy transfers. The singular behavior for low $\vec{q}$ for $\hbar\omega = 0.2$ eV corresponds to the anomalous behavior shown in Fig. 1. The qualitative behavior of the real part is again consistent with the calculations of the static susceptibility due to Hebborn and March[7] and Julien *et al.*[26]

C. Exchange-enhanced susceptibility function and comparison with measured $S(\vec{q},\omega)$

In this section we compare our results with the neutron scattering measurements. For a paramagnetic system the reduced susceptibility is related to the scattering function $S(\vec{q},\omega)$ via

$$S(\vec{q},\omega) = -(\hbar/\pi g^2\mu_B^2)(1-e^{-\hbar\omega\beta'})^{-1}\mathrm{Im}\bar{\chi}(\vec{q},\omega), \quad (64)$$

where $\mathrm{Im}\bar{\chi}(\vec{q},\omega)$ is the imaginary exchange enhanced reduced susceptibility function and $\beta' = 1/k_BT$. In the random-phase approximation the exchange-enhanced reduced susceptibility can be written[27]

$$\mathrm{Im}\bar{\chi}(\vec{q},\omega) = \mathrm{Im}\bar{\chi}^0(\vec{q},\omega)/\{[1-(2/g^2\mu_B^2)I_{\mathrm{eff}}(\vec{q})\,\mathrm{Re}\bar{\chi}^0(\vec{q},\omega)]^2 + [(2/g^2\mu_B^2)I_{\mathrm{eff}}(\vec{q})\,\mathrm{Im}\bar{\chi}^0(\vec{q},\omega)]^2\}. \quad (65)$$

Allan *et al.*[10] reviewed the static and momentum-dependent values of I_{eff}. They also extracted out the momentum dependence of I_{eff} while comparing their results with neutron scattering measurements. But all those expressions for I_{eff} are valid only for a small region of $\vec{q}$. Singwi *et al.*[28] calculated $I_{\mathrm{eff}}(\vec{q})$ self-consistently in the free-electron approximation which is hardly adoptable for *d* electrons. In view of these uncertainties, the present calculations for the exchange-enhanced susceptibility function are performed for the static values of I_{eff}. First, $S(\vec{q},\omega)$ is calculated from the unenhanced reduced susceptibility function which is shown in Fig. 3 by a dash-dot line. The results for $S(\vec{q},\omega)$ obtained from the unenhanced reduced susceptibility function due to Allan *et al.*[10] are also shown in Fig. 3. The qualitative behavior of $S(\vec{q},\omega)$ is consistent with the calculation of Allan *et al.*[20] The value of I_{eff} is then varied to get the best fit with the experimental results. A reasonably good agreement is obtained for $I_{\mathrm{eff}} = 0.29$ eV which is close to the value 0.32 eV suggested by Allan *et al.*[10]

IV. DISCUSSION

In the evaluation of the dynamical spin susceptibility in this paper we use a model band structure which has reduced the computational efforts as it made possible to evaluate many expressions analytically. In principle one should use a wave function for *s* electrons which is orthogonal to core and *d* wave functions. An orthogonalized plane wave is a suitable choice, but it has been found that orthogonalization corrections are very small[29] and therefore the use of a simple plane wave for *s* electrons is justified. The *d* bands are in fact flat near the zone boundary, which gives the structural features in the susceptibility function. The parabolic-band approximation is, however, crucial and does not reproduce the exact structural features. The temperature dependence of the susceptibility function is also neglected in our cal-

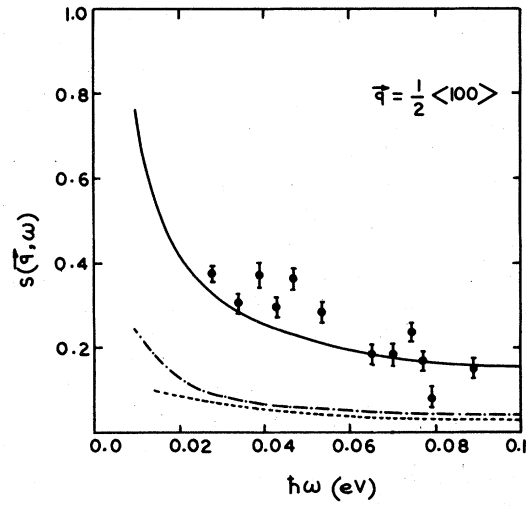

FIG. 3. $S(\vec{q},\omega)$ vs $\hbar\omega$ for paramagnetic nickel. Dash-dot line shows $S(\vec{q},\omega)$ as calculated from the unenhanced reduced susceptibility function and the solid line represents $S(\vec{q},\omega)$ as calculated from the enhanced reduced susceptibility function. Dotted line shows $S(\vec{q},\omega)$ as calculated from the unenhanced reduced susceptibility function by Allan *et al.* (Ref. 10) and the filled circles denote the experimental values.

culations. The contribution due to the orbital susceptibility which is assumed to be small as already pointed out by Hebborn and March,[7] is again neglected while comparing our results with the experimental data. Our results obtained with considerably reduced computational effort, agree well with the calculations of Allan *et al.*[10] where a realistic band-structure calculation is used.

ACKNOWLEDGMENTS

The authors wish to express their thanks to Professor S. K. Joshi and K. N. Pathak for fruitful and informative discussions and Professor H. S. Hans for encouragement. The financial support from the Council of Scientific and Industrial Research, New Delhi, is also acknowledged.

*Present address: Laboratoire de Chimie Physique, Bâtiment 350, Avenue Jean Perrin 91405 Orsay, France.

[1]R. Abe, Progr. Theor. Phys. (Kyoto) 29, 23 (1963).
[2]A. G. Samoilovich and E. Ya. Rabinovich, Fiz. Tverd. Tela 5, 778 (1963) [Sov. Phys.-Solid State 5, 567 (1963)].
[3]M. L. Glasser, Phys. Rev. 134, A1296 (1964).
[4]K. Yamaji and R. Kubo, J. Phys. Soc. Jpn. 25, 330 (1968).
[5]P. K. Misra and L. M. Roth, Phys. Rev. 177, 1089 (1969).
[6]R. D. Lowde and C. G. Windsor, Adv. Phys. 19, 813 (1970), and the references therein.
[7]J. E. Hebborn and N. H. March, Adv. Phys. 19, 175 (1970), and the references therein.
[8]S. Doniach, Proc. Phys. Soc. 91, 86 (1967).
[9]T. Izuyama, D. J. Kim, and R. Kubo, J. Phys. Soc. Jpn. 18, 1025 (1963).
[10]G. Allan, W. M. Lomer, R. D. Lowde, and C. G. Windsor, Phys. Rev. Lett. 20, 933 (1968).
[11]H. Yamada and M. J. Shimizu, J. Phys. Soc. Jpn. 22, 1404 (1967); 25, 1001 (1968).
[12]J. B. Sokoloff, Phys. Rev. 180, 613 (1969); 185, 770 (1969); 185, 783 (1969).
[13]R. P. Gupta and S. K. Sinha, Phys. Rev. B 3, 2401 (1971).
[14]J. F. Cooks and R. F. Wood, Phys. Rev. B 7, 893 (1973), A. I. P. Conf. Proc. 10, 1218 (1973).
[15]R. J. Rath and A. J. Freeman, Phys. Rev. B 11, 2109 (1975).
[16]Natthi Singh, J. Singh, and S. Prakash, Phys. Rev. B 12, 1076 (1975).
[17]Natthi Singh and S. Prakash, Phys. Rev. B 8, 5532 (1972); 12, 1600 (1975).
[18]Natthi Singh, J. Singh, and S. Prakash, Phys. Rev. B 12, 5415 (1975).
[19]S. Prakash and S. K. Joshi, Phys. Rev. B 5, 2880 (1972).
[20]S. Prakash and N. Singh, Physica (Utr.) 78, 273 (1974).
[21]S. Prakash and S. K. Joshi, Phys. Rev. B 2, 915 (1970).
[22]E. Hayashi and M. Shimizu, J. Phys. Soc. Jpn. 26, 1396 (1969).
[23]A. Czachor, Phys. Rev. B 9, 3357 (1974).
[24]R. E. Watson, M.I.T. Solid State and Molecular Theory group Technical Report No. 12 (1952) (unpublished).
[25]J. Singh, N. Singh, and S. Prakash, Phys. Rev. B 12, 3159 (1975); 12, 3166 (1975).
[26]R. Jullien, M. T. Beal-Monod, and B. Coqblin, Phys. Rev. B 9, 1441 (1974).
[27]S. Doniach, in *Rendiconti della scuola Internazionale de Fisica "Enrico Fermi" XXXVII Corso*, edited by W. Marshall (Academic, New York, 1967), p. 319.
[28]K. S. Singwi, A. Sjölander, M. P. Tosi, and R. H. Land, Phys. Rev. B 1, 1044 (1970).
[29]W. R. Hanke, Phys. Rev. B 8, 4558–96 (1973).