# Temperature dependence of dynamical spin susceptibility of transition metals

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A formalism is developed for the temperature-dependent dynamical spin susceptibility using a multiband scheme for transition metals. The imaginary part of the dynamical spin susceptibility is derived using the temperature-dependent Fermi distribution function. Kramers-Kronig relations are used to calculate the real part of the susceptibility function. The limiting cases are derived and discussed in the light of available theoretical results. The detailed calculations are carried out for ferromagnetic and paramagnetic nickel, palladium, and platinum for various values of momentum and energy transfer. It is found that the susceptibility decreases with the increase of temperature and the peaks at small values of momentum transfer are broadened and show a decrease in magnitude.

### I. INTRODUCTION

Frequency- and wave-vector-dependent susceptibility is an important quantity in the study of many physical properties such as resistivity, specific heat, Lorentz number, thermopower, and ferromagnetism, etc. of a system. The various spin susceptibility calculations may be classified into two model groups: the localized spin model in which electrons are supposed to be localized at each atom and the itinerant electron model in which the electrons are represented by Bloch waves. Many authors<sup>1-6</sup> have calculated the time-independent spin susceptibility which does not explain satisfactorily the results for specific heat and resistivity. Rivier and Zuckermann<sup>7</sup> emphasized the importance of temperature variation of spin susceptibility of metals and alloys in order to explain these physical properties. Doniach and Engelsberg<sup>8</sup> and Shimizu et al.<sup>9</sup> have shown that the temperaturedependent static susceptibility can explain the specific-heat data at finite temperatures.

It is well know that the resistivity shows  $T^2$  law at low temperatures and T law at high temperatures. Mills and Lederer<sup>3</sup> calculated the resisitivity of dband metals, taking into account Izuyama et al.<sup>2</sup> dynamical transverse susceptibility at low and high temperatures in the itinerant electron model. It has been observed that for nearly magnetic metals and alloys<sup>10</sup> at high temperature, the resistivity departs from T law and has negative slope. The first improvement was done by Kaiser and Doniach<sup>11</sup> who used a low-temperature expansion for the spin susceptibility in the calculation of resistivity and then Rivier and Zlatic<sup>12</sup> calculated the resistivity in the high-temperature limit. Jullien et al.<sup>13</sup> explained the essential features of paramagnon resistivity of nearly magnetic metals using full temperature-dependent dynamical spin susceptibility  $\chi(\vec{q}, \omega, T)$ .

Izuyama et al.<sup>2</sup> have shown that the itinerant electron model can give a satisfactory description of the neutron scattering data. Moreover, the large specific heat and the fractional Bohr magnetons per atom suggest that the itinerant electron model is advantageous for transition metals. In this paper we present the formalism and calculations for the temperaturedependent dynamical spin susceptibility for various values of energy and momentum transfer for transition metals. The model band structure<sup>14</sup> with fivefold degenerate d band and an s band is used in the calculation of susceptibility for Ni, Pd, and Pt. The susceptibility is found to decrease with the increase of temperature and the peaks at small momentum transfer are found to broaden. In the present model the ferromagnetism in nickel is found in accordance with the conclusion of Izuyama et al.<sup>2</sup> The plan of the paper is as follows:

The formalism for susceptibility and the model band structure are presented in Sec. II; the calculations and results are presented in Sec. III and these are discussed in Sec. IV.

### **II. THEORY**

The general expression for the temperaturedependent dynamical spin susceptibility is given as<sup>15</sup>.

$$\chi^{0}(\vec{\mathbf{q}},\,\omega,T) = \sum_{\sigma\sigma'} \chi^{0}_{\sigma\sigma'}(\vec{\mathbf{q}},\,\omega,T) \quad , \tag{1}$$

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where

$$\chi^{0}_{\sigma\sigma'}(\vec{q},\omega,T) = \frac{1}{4}g^{2}\mu^{2}_{B}\lim_{\epsilon \to 0} \sum_{\vec{k}'} \sum_{lm \atop l'm'} \frac{f(E_{lm\sigma}(\vec{k}),T) - f(E_{l'm'\sigma'}(\vec{k}+\vec{q}),T)}{E_{l'm'\sigma'}(\vec{k}+\vec{q}) - E_{lm\sigma}(\vec{k}) - \hbar\omega - i\epsilon} |\langle \Psi_{lm\sigma}(\vec{k})|e^{-i\vec{q}\cdot\vec{\tau}}|\Psi_{l'm'\sigma'}(\vec{k}+\vec{q})\rangle|^{2}$$

$$(2)$$

Here *l*, *m*, and  $\sigma$  are the oribital, magnetic, and spin quantum numbers and also act as band indices.  $E_{lm\sigma}(\vec{k})$  is the energy eigenvalue for the Bloch function  $\Psi_{lm\sigma}(\vec{k})$  and  $f(E_{lm\sigma}(\vec{k}),T)$  is the Fermi distribution function. It will be too difficult to evaluate Eq. (2) accounting rigorously the detailed bandstructure calculations. However, to bring out the salient features of temperature-dependent susceptibility and for the sake of simplicity we adopt a model band structure proposed by Prakash and Joshi<sup>14</sup> and use it to calculate the dynamical spin susceptibility. In the following we discuss separately the model band structure briefly for completeness and the calculations of dynamical spin susceptibility.

### A. Model band structure

In the model band structure the s-d interaction is neglected and the noninteracting s and d bands are constructed along the principal symmetry directions using the results of the detailed band-structure calculations and group-theoretical considerations. The Houston averaging method is used to construct the isotropic noninteracting bands. A schematic example for paramagnetic Ni, Pd, and Pt is shown in Fig. 1. The detailed calculations are given in references [Ni,<sup>14</sup> Pd,<sup>16</sup> and Pt (Ref. 16)]. The effective masses, Fermi momenta, and the parameters for the radial wave functions are adopted from the same references. In this model the conduction electrons of the transition metal are characterized to be of s and dtype and these are assumed to occupy the noninteracting s and d band.

In the noninteracting isotropic band models for paramagnetic Ni, Pd, and Pt, one s and one d subband are found partially filled and other d subbands completely filled. Under the action of the applied field the electrons will readjust themselves through the following transitions: (i) from partially filled s band to partially filled s band (s-s), (ii) from filled and partially filled d subbands to partially filled d subbands (d-d), (iii) from filled and partially filled d subbands to partially filled s band (d-s), and (iv) from partially filled s band to partially filled d subbands (s-d). These intraband and interband contributions are computed by Prakash and Joshi<sup>14</sup> and Singh et al.<sup>16</sup> It is found that the interband contributions are unimportant compared to intraband contributions in the small- $\vec{q}$  region while both are comparable in the large- $\vec{q}$  region. Since we restrict our calculations only in the small- $\vec{q}$  region we therefore explicitly consider the intraband transitions and ignore the interband contributions completely. In the intraband transitions only the partially filled s and d subbands contribute. The chemical potential is the same while the effective masses and Fermi momenta for s and d subbands are different because of the different slopes and different points of intersection with Fermi energy.

Similarly an isotropic noninteracting spin band model is constructed for ferromagnetic Ni. From majority and minority noninteracting spin bands it is found that the intraband transitions will take place only in the partially filled majority and minority spin sbands and in the partially filled minority spin d sub-



FIG. 1. Schematic isotropic noninteracting band model for Ni, Pd, and Pt.

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bands. The Fermi momenta and effective masses for 's and d subbands are calculated with the help of Fermi energy.

B. Calculation of 
$$\chi^0_{\sigma\sigma'}(\vec{q}, \omega, T)$$

In the above isotropic noninteracting band model, Eqs. (1) and (2) simplify as

$$\chi^{0}(\vec{q},\omega,T) = \sum_{\sigma} \chi^{0}_{\sigma}(\vec{q},\omega,T) \quad , \tag{3}$$

where

$$\chi^{0}_{\sigma}(\vec{q},\omega,T) = \frac{1}{4}g^{2}\mu^{2}_{B}\lim_{\epsilon \to 0} \sum_{\vec{k}} \sum_{lm} \frac{f(E_{lm\sigma}(\vec{k}),T) - f(E_{lm\sigma}(\vec{k}+\vec{q}),T)}{E_{lm\sigma}(\vec{k}+\vec{q}) - E_{lm\sigma}(\vec{k}) - \hbar\omega - i\epsilon} |\langle \Psi_{lm\sigma}(\vec{k})|e^{-i\vec{q}\cdot\vec{r}}|\Psi_{lm\sigma}(\vec{k}+\vec{q})\rangle|^{2} .$$
(4)

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We represent the s electrons by plane waves and the d electrons by the simple tight-binding wave functions which simplify the above expressions in the following form:

$$\chi^{0}_{\sigma}(\vec{q},\omega,T) = \chi^{0}_{s\sigma}(\vec{q},\omega,T) + \sum_{m} \chi^{0}_{dm\sigma}(\vec{q},\omega,T) , \qquad (5)$$

where

$$\chi^{0}_{s\sigma}(\vec{\mathbf{q}},\omega,T) = \frac{1}{4}g^{2}\mu^{2}_{B}\lim_{\epsilon \to 0} \sum_{\vec{\mathbf{k}}} \frac{f(E_{s\sigma}(\vec{\mathbf{k}}),T) - f(E_{s\sigma}(\vec{\mathbf{k}}+\vec{\mathbf{q}}),T)}{E_{s\sigma}(\vec{\mathbf{k}}+\vec{\mathbf{q}}) - E_{s\sigma}(\vec{\mathbf{k}}) - \hbar\omega - i\epsilon}$$
(6)

and

$$\chi^{0}_{dm\sigma}(\vec{q},\omega,T) = \frac{1}{4}g^{2}\mu^{2}_{B}\lim_{\epsilon \to 0} \sum_{\vec{k}} \frac{f(E_{dm\sigma}(\vec{k}),T) - f(E_{dm\sigma}(\vec{k}+\vec{q}),T)}{E_{dm\sigma}(\vec{k}+\vec{q}) - E_{dm\sigma}(\vec{k}) - \hbar\omega - i\epsilon} |\Delta^{(q)}_{dm,dm}|^{2} .$$
<sup>(7)</sup>

 $\Delta_{dm,dm}^{(q)}$  are the overlap matrix elements whose explicit expressions are given by Prakash and Joshi.<sup>14</sup> Different *d* subbands usually have different radial functions which are more extended for the lower (bonding) *d* bands and more contracted for the upper (antibonding) *d* subbands. This would lead to differences in the matrix elements for different values of *m*. However for simplicity, we ignore this difference and take an arithmetic mean over all *m* values giving equal weight to each component.<sup>6</sup> This leads to

$$\Delta_{dm,dm}^{(q)} = I_0(q) = \int_0^\infty j^0(qr) R_{nd}^2(r) dr \quad , \tag{8}$$

where  $j^0(qr)$  is the zeroth-order spherical Bessel function and  $R_{nd}$  is the radial *d*-wave function for the principal quantum number *n*. Using Eq. (8) in Eq. (7), one can write

$$\chi^{0}_{dm\sigma}(\vec{q},\omega,T) = \frac{1}{2}g^{2}\mu^{2}_{B}\lim_{\epsilon \to 0} \sum_{\vec{k}} I^{0}_{0}(q) \frac{f(E_{dm\sigma}(\vec{k}),T) - f(E_{dm\sigma}(\vec{k}+\vec{q}),T)}{E_{dm\sigma}(\vec{k}+\vec{q}) - E_{dm\sigma}(\vec{k}) - \hbar\omega - i\epsilon}$$
(9)

Here  $dm\sigma$  stands for *m* th *d* subband with spin  $\sigma$ . From Eq. (8) one can easily see that  $I_0(q) \rightarrow 1$  as  $q \rightarrow 0$ . Therefore, for  $q = \omega = T = 0$ , both  $\chi^0_{s\sigma}(\vec{q}, \omega, T)$  and  $\chi^0_{dm\sigma}(\vec{q}, \omega, T)$  reduce to one-half of the density of states, i.e.,

$$\chi^{0}_{s\sigma}(0,0,0) = \frac{m_{s\sigma}k_{F_{s\sigma}}}{2\pi^{2}\hbar^{2}} \Omega = \frac{1}{2}N(E^{F}_{s\sigma}(0)) , \qquad \chi^{0}_{dm\sigma}(0,0,0) = \frac{m_{dm\sigma}k_{F_{dm\sigma}}}{2\pi^{2}\hbar^{2}} \Omega = \frac{1}{2}N(E^{F}_{dm\sigma}(0)) ,$$

$$N(E^{F}_{\sigma}(0)) = N(E^{F}_{s\sigma}(0)) + N(E^{F}_{dm\sigma}(0)) .$$
(10)

 $m_{s\sigma}$  and  $m_{dm\sigma}$  are the effective masses for the s band and mth d subband.  $k_{F_{s\sigma}}$  and  $k_{F_{dm\sigma}}$  are the corresponding Fermi momenta.  $\Omega$  is the total volume of the crystal and  $N(E_{\sigma}^{F}(0))$  is the total density of states. Using the identity

$$\lim_{\epsilon \to 0} \frac{1}{\chi \pm i\epsilon} = \frac{1}{\chi} \mp i\pi\delta(\chi)$$
(11)

the imaginary part of Eq. (6) can be written

$$\operatorname{Im}\chi^{0}_{s\sigma}(\vec{q},\omega,T) = \frac{1}{4}g^{2}\mu^{2}_{B}\pi \sum_{\vec{k}} \left[ f\left( E_{s\sigma}(\vec{k}),T\right) - f\left( E_{s\sigma}(\vec{k}+\vec{q}),T\right) \right] \delta\left( E_{s\sigma}(\vec{k}+\vec{q}) - E_{s\sigma}(\vec{k}) - \hbar\omega \right) \quad . \tag{12}$$

Replacing the summation over  $\vec{k}$  by an integration and solving the  $\delta$  function integral in the parabolic band approximation the above equation reduces as

$$\operatorname{Im} \chi^{0}_{s\sigma}(\vec{q},\omega,T) = \frac{g^{2} \mu_{B}^{2} m_{s\sigma} \Omega}{16\pi \hbar^{2} q} \int_{|m_{s\sigma}\omega/\hbar q - q/2|}^{\infty} \left[ f(E_{s\sigma}(\vec{k}),T) - f(E_{s\sigma}(\vec{k}+\vec{q}),T) \right] k \, dk \quad . \tag{13}$$

Solving analytically the Eq. (13) and putting  $\hbar = k_B = 1$ , the imaginary part of the reduced spin susceptibility  $\bar{\chi}^0_{s\sigma}(\vec{q}, \omega, T) = \chi^0_{s\sigma}(\vec{q}, \omega, T)/\chi^0_{s\sigma}(0, 0, 0)$  becomes

$$\operatorname{Im}\overline{\chi}^{0}_{s\sigma}(\vec{q},\omega,T) = \frac{1}{2}\pi \frac{m_{s\sigma}T}{qk_{F_{s\sigma}}} \ln \left| \frac{e^{\omega/T} + \exp[\epsilon_{s\sigma}(\vec{q},\omega,T)/T]}{1 + \exp[\epsilon_{s\sigma}(\vec{q},\omega,T)/T]} \right|,$$
(14)

where

$$\epsilon_{s\sigma}(\vec{q},\omega,T) = \frac{1}{2m_{s\sigma}} \left( \frac{m_{s\sigma}\omega}{q} + \frac{q}{2} \right)^2 - E_{s\sigma}^F(T) \quad .$$
(15)

In exactly the same manner the contribution to the spin susceptibility due to the d subbands is calculated which is given as

$$\operatorname{Im}\overline{\chi}^{0}_{dm\,\sigma}(\vec{q},\,\omega,T) = \frac{\pi}{2} \frac{m_{dm\,\sigma}T}{qk_{F_{dm\,\sigma}}} I_{0}^{2}(q) \ln \left| \frac{e^{\omega/T} + \exp[\epsilon_{dm\,\sigma}(\vec{q},\,\omega,T)/T]}{1 + \exp[\epsilon_{dm\,\sigma}(\vec{q},\,\omega,T)/T]} \right| , \tag{16}$$

where

$$\epsilon_{dm\sigma}(\vec{q},\omega,T) = \frac{1}{2m_{dm\sigma}} \left( \frac{m_{dm\sigma}\omega}{q} + \frac{q}{2} \right)^2 - E_{dm\sigma}^F(T) \quad .$$
(17)

The real part of the spin susceptibility  $\overline{\chi}^0(\overline{q}, \omega, T)$  is obtained using Kramers-Kronig relation

$$\operatorname{Re}\overline{\chi}^{0}(\vec{q},\omega,T) = \frac{2}{\pi} \int_{0}^{\infty} \frac{\omega' \operatorname{Im}\overline{\chi}^{0}(\vec{q},\omega,T)}{(\omega')^{2} - \omega^{2}} d\omega' \quad .$$
(18)

From Eqs. (14), (16), and (18) the reduced static susceptibility  $\overline{\chi}^0(\vec{q}, 0, T)$  is obtained. At T = 0 the static susceptibility for s and d electrons simplifies as

$$\overline{\chi}^{0}_{s\sigma}(\vec{q},0,0) = \frac{1}{2} \left[ 1 + \frac{4k_{F_{s\sigma}}^{2} - q^{2}}{4k_{F_{s\sigma}}q} \ln \left| \frac{2k_{F_{s\sigma}} + q}{2k_{F_{s\sigma}} - q} \right| \right] , \qquad (19)$$

and

$$\bar{\chi}^{0}_{dm\,\sigma}(\vec{q},0,0) = \frac{1}{2}I^{2}_{0}(q) \left( 1 + \frac{4k^{2}_{F_{dm\,\sigma}} - q^{2}}{4k_{F_{dm\,\sigma}}q} \ln \left| \frac{2k_{F_{dm\,\sigma}} + q}{2k_{F_{dm\,\sigma}} - q} \right| \right)$$
(20)

These are Lindhard functions<sup>17</sup> for s and d subbands. The only difference between s- and d-band contributions is that in the latter part the effect of overlap matrix elements is included, which decreases the d-band contribution much faster than that due to s band.

It should be mentioned that the parabolic band is a good representation for s electrons. For d electrons this representation is only good when the band is slightly populated at the bottom. In the present calculation, the d

band is more than half filled therefore the parabolic band is a poor representation. However, this approxiamtion greatly simplifies the calculations and therefore we adopt it while evaluating d-band contributions. Certainly it will overestimate the d-band contributions but we expect the qualitative features due to d bands to be included in it.

From Eqs. (16) and (18) the real part of the dynamical susceptibility at T = 0 is obtained as follows:

$$\operatorname{Re} \overline{\chi}_{dm\sigma}^{0}(\overline{q},\omega,0) = \frac{1}{2} I_{0}^{2}(q) + \frac{1}{4} I_{0}^{2}(q) \frac{k_{F_{dm\sigma}}}{q} \times \left\{ \left[ 1 - \left( \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} + \frac{q}{2k_{F_{dm\sigma}}} \right)^{2} \right] \ln \left| \left( 1 + \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} + \frac{q}{2k_{F_{dm\sigma}}} \right) / \left( 1 - \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} - \frac{q}{2k_{F_{dm\sigma}}} \right) \right| - \left[ 1 - \left( \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} - \frac{q}{2k_{F_{dm\sigma}}} \right)^{2} \right] \ln \left| \left( 1 + \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} - \frac{q}{2k_{F_{dm\sigma}}} \right) / \left( 1 - \frac{m_{dm\sigma}\omega}{qk_{F_{dm\sigma}}} + \frac{q}{2k_{F_{dm\sigma}}} \right) \right| \right\}, \quad (21)$$

where we have used

$$\operatorname{Im}\overline{\chi}^{0}_{dm\,\sigma}(\vec{q},\,\omega,\,0) = \frac{1}{2}\pi\frac{m_{dm\,\sigma}\omega}{ak_{F_{dm\,\sigma}}}I_{0}^{2}(q) \quad \text{if} \quad \left|\frac{m_{dm\,\sigma}\omega}{qk_{F_{dm\,\sigma}}}\right| < \left|1 - \frac{q}{2k_{F_{dm\,\sigma}}}\right| \quad \text{and} \quad \frac{q}{2k_{F_{dm\,\sigma}}} < 2$$
$$= \frac{1}{4}\pi\frac{k_{F_{dm\,\sigma}}}{q} \left[1 - \left(\frac{m_{dm\,\sigma}\omega}{qk_{F_{dm\,\sigma}}} - \frac{q}{2k_{F_{dm\,\sigma}}}\right)^{2}\right]I_{0}^{2}(q) \quad \text{if} \quad \left|1 - \frac{q}{2k_{F_{dm\,\sigma}}}\right| < \left|\frac{m_{dm\,\sigma}\omega}{qk_{F_{dm\,\sigma}}}\right| < \left|1 + \frac{q}{2k_{F_{dm\,\sigma}}}\right|$$

$$=0$$
 otherwise ,

which is obtained by simplifying Eq. (16). Equation (21) is just the Lindhard function<sup>17</sup> for the dynamical spin susceptibility multiplied by the factor  $I_0^2(q)$ . The expression for  $\operatorname{Re} \overline{\chi}_{s\sigma}^0(\overline{q}, \omega, 0)$  is obtained if we put  $I_0(q) = 1$  and replace the effective mass and Fermi momentum of the *d* electrons (subband) by that of *s* electrons (band).

## **III. CALCULATIONS AND RESULTS**

Evidently, from Eqs. (14)-(17) the temperature dependence in the dynamical susceptibility is introduced through the chemical potential  $E^{F}(T)$ . The energy eigenvalues and the eigenfunctions are treated as temperature independent. The temperature dependence of the chemical potential is calculated with the help of conservation law.<sup>13, 18</sup>

$$2\sum_{n}\sum_{k}f(E_{n}(k),T) = N , \qquad (23)$$

where N is the total number of conduction electrons per atom. In the present model  $N = N_s + N_d$  where  $N_s$  and  $N_d$  are the total number of s and d electrons distributed in the partially filled s and d subbands. We use the atomic configuration  $(nd)^{9.4}[(n + 1)]^{0.6}$ wherein n = 3, 4, and 5 for Ni, Pd, and Pt, respectively. In the present model band scheme described earlier, the effective electronic charges -1.4e and -0.6e per atom are taken in the partially filled d and s band, respectively.

With the change of temperature the chemical potential changes and it is quite likely that the effective electronic charge per atom in the s and d subbands may change. This change may arise due to interband s-d and d-s transitions and s-d hybridization which has not been included explicitly in the present model. As regards to the interband transitions, it has been shown explicitly in the previous calculations<sup>5, 6</sup> that the interband part is negligible compared to the intraband part, therefore the contribution of the interband part towards the change in the effective electronic charge per atom in the s and d subbands will be negligible. It has been pointed out by Chan and Young,<sup>19</sup> and Yamada and Shimizu<sup>20</sup> that the interband part gives rise to Stoner-type excitations and is presumably not important in the low- $\vec{q}$  region. According to Clark and Young<sup>21</sup> for the intraband transitions, if

(22)

the particle and hole pair start off in the same band n, they will end up in the same band n' (where n' may not necessarily be equal to n). According to this prescription as the temperature varies, the number of electrons (during the intraband transitions) remain conserved individually in a band, though the character of the band may change from s to d type and vice versa. However, Bringer and Lustfeld<sup>22</sup> have shown that in the case of mixed-valence crystals the number of s and f electrons is not conserved individually because of the large s-f mixing. In these compounds, a certain fraction of rare-earth ions transfer electrons to the conduction band.

Using the parabolic-band approximation for s and d electrons Eq. (23) simplifies as

$$\sum_{n} \int_{0}^{\infty} \frac{E(\vec{k}) dE(\vec{k})}{\left[1 + \exp\left(\frac{E(\vec{k}) - E^{F}(T)}{T}\right)\right]} = \frac{2}{3} \left[E^{F}(0)\right]^{3/2} ,$$
(24)

where  $E^F(0) (=k_F^2/2m = T_F)$  is the Fermi temperature for electrons. Equation (24) gives a unique relation of  $E_{i\sigma}^F(T)/E_{i\sigma}^F(0)$  with  $T/T_{F_{i\sigma}}$  which is obtained through numerical calculations and these results are shown in Fig. 2 (*i* is index for *s* and *d* subbands). Our results are similar to those obtained by Jullien *et al.*<sup>13</sup> Since the relative chemical potential is the same for both the *s* and *d* electrons therefore this behavior is valid for both the *s* and *d* electrons.

For ferromagnetic Ni, the temperature dependence of the spin splitting is not considered explicitly. The



FIG. 2. Temperature variation of chemical potential for s band, i.e.,  $E_{I\sigma}^{F}(T)/E_{I\sigma}^{F}(0)$  vs  $T/T_{F_{i\sigma}}$ . The quantities are same as defined in the text. The index *i* stands for s and d subbands.

temperature variation of chemical potential which explicitly includes the temperature variation of spin splitting is studied using Eqs. (23) and (24). Again the number of electrons per atom in each subband is kept fixed to be consistent with the noninteracting spin band model. The limitations of the model will not allow us to study ferromagnetic-paramagnetic phase transition because the electronic band structure for paramagnetic and ferromagnetic phases are adopted separately.

The reduced static and dynamical susceptibilities as a function of  $\vec{q}$ ,  $\omega$ , and T are calculated for paramagnetic and ferromagnetic Ni, Pd, and Pt. The required parameters are adopted from the Refs. 14 and 23. To study the relative contributions of the s and d subbands the results for the imaginary part of the reduced spin susceptibility as a function of  $\omega$  for fixed values of q and T for ferromagnetic Ni are shown in Fig. 3. We find that in the vicinity of  $\omega = 0$ , the dband contribution dominates and shows two peaks. For larger values of  $\omega$ , the s-band contribution dominates and shows a strong peak. Both the contributions fall sharply to zero for finite values of  $\omega$ .

The detailed results of the imaginary part as a function of  $\omega$  for Ni, Pd, and Pt for various values of  $\vec{q}$  and  $T/T_{F_{i\sigma}}$  are shown in Fig. 4. For ferromagnetic Ni [Figs. 4(a) and 4(b)] we find that there are two peaks for small values of  $\omega$  for the wave vectors q = 0.05 and 0.25. These distinct peaks may be ascribed to the leading contributions of the two partially filled minority spin d subbands. The broad peak at



FIG. 3.  $\text{Im}\overline{\chi}^0(\vec{q}, \omega, T)$  vs  $\omega$  for ferromagnetic Ni at  $T/T_{F_{i\sigma}} = 0$  and q = 0.25. The s- and d-band contributions are shown separately. In  $T_{F_{i\sigma}}$ , *i* stands for both the s and d bands.



FIG. 4.  $\text{Im}\overline{X}^0(\vec{q}, \omega, T)$  vs  $\omega$ . (a) and (b) show the results for ferromagnetic Ni for q = 0.05 and 0.25, respectively. Curves labeled 1, 2, 3, 4 correspond, respectively, to  $T/T_{F_{i\sigma}} = 0.0, 0.2, 0.4, 1.2$ . (c) and (d) show results for paramagnetic Ni, (e) and (f) for Pd, and (g) and (h) for Pt. The description for Ni, Pd, and Pt is the same as that for ferromagnetic Ni.

large values of energy transfer is due to the contribution of the partially filled majority and minority s bands. These peaks get separated with the increase of q. From curves 1 to 4, we see that as the temperature goes on increasing the peaks get more and more broadened. In curve 4, the structural features almost vanish and the two peaks at small values of  $\omega$ merge into one. Therefore, we may conclude that the effect of increasing the temperature is to destroy the spin waves and this leads to the paramagnetic phase, although the continuous ferro-para phase transition is not possible in the present model.

For ferromagnetic Ni [Figs. 4(c) and 4(d)] there is one peak at small values of  $\omega$  and another peak at large values of  $\omega$  for both the values of momentum transfer (q = 0.05 and 0.25). These correspond to the contributions from one partially filled d subband and from one s band, respectively. The variation with T and  $\omega$  is the same as that found for ferromagnetic Ni. Similar results are also found for Pd [Figs. 4(e) and 4(f)] and Pt [Figs. 4(g) and 4(h)]. In general with increasing temperature the paramagnons are destroyed as the characteristic peaks became broadened and decrease in magnitude at high temperatures. We find that  $\text{Im}\overline{\chi}^0(\vec{q}, \omega, T)$  becomes almost smooth and energy dependence becomes weak at high temperatures. As the real part of the susceptibility is evaluated directly from the imaginary part so

these structural features will also be reflected in the real part.

The variation of the static reduced spin susceptibility as a function of momentum transfer and temperature is also studied. The expressions for the imaginary part of the susceptibility function vanish in the  $\omega \rightarrow 0$  limit and we are left with the expressions only for the real part. The calculated results for Ni, Pd, and Pt are shown in Fig. 5. The dashed lines in Fig. 5 represent the results in the high-temperature limit (Curie limit). For ferromagnetic Ni we note that  $\overline{\chi}^{\circ}(\overline{q}, 0, T)$  decreases rapidly for small values of  $\overline{q}$ and after that the decrement is smooth and structureless. The structural features at small values of  $\vec{q}$  are due to the dominating contribution of d electrons, which vanishes rapidly for large values of  $\vec{q}$ . As the temperature increases the magnitude of the susceptibility decreases and the peaked behavior at small  $\vec{q}$ becomes diffused. Evidently, from Fig. 5(b) the susceptibility decreases in magnitude with the increase of  $\vec{q}$  and T. The results for paramagnetic Ni are shown in Figs. 5(c) and 5(d), for Pd in Figs. 5(e) and 5(f), and for Pt in Figs. 5(g) and 5(h).

While comparing the results for ferromagnetic and paramagnetic Ni, Pd, and Pt we find that the magnitude of the static temperature-dependent susceptibili-



FIG. 5. Variation of reduced static susceptibility  $\overline{\chi}^0(\overline{q}, 0, T)$  with  $\overline{q}$  and temperature. (a) shows  $\overline{\chi}^0(\overline{q}, 0, T)$  vs  $\overline{q}$  and (b) shows  $\overline{\chi}^0(\overline{q}, 0, T)$  vs  $T/T_{F_{i\sigma}}$  for ferromagnetic Ni. In (a) curves labeled 1,2,3,4 correspond, respectively, to  $T/T_{F_{i\sigma}} = 0.2, 0.4, 0.8, 1.0$  while in (b) curves labeled 1, 2, 3, 4 correspond, respectively, to q = 0.05, 0.25, 0.65, 1.05. (c) and (d) show the static susceptibility for paramagnetic Ni, (e) and (f) for Pd, and (g) and (h) for Pt. The description is the same as that of (a) and (b). The dashed lines show the asymptotic limit with respect to temperature.

ty for ferromagnetic Ni is much larger than that for paramagnetic Ni, Pd, and Pt because the d-band contribution which depends directly upon the effective masses, is larger in ferromagnetic Ni than in paramagnetic Ni, Pd, and Pt. The d-band contribution however decreases faster than the s-band contribution with increasing values of  $\vec{q}$ . The susceptibility decreases with the increase of temperature and obeys the simple Curie law at high temperatures for which the results are shown by dashed lines. The susceptibility is different for different  $\vec{q}$  at low temperatures but approaches the same value at high temperatures as pointed out earlier. The decrease of susceptibility with temperature is consistent with the calculations of Shimizu *et al.*<sup>24</sup> Re $\bar{\chi}^0_{s\sigma}(\vec{q}, 0, 0)$  and Re $\bar{\chi}^0_{dm\sigma}(\vec{q}, 0, 0)$ show anomalies at  $q = 2k_{F_{s\sigma}}$  and  $q = 2k_{F_{dm\sigma}}$  at T = 0but these anomalies disappear at finite temperatures. The qualitative features of the present results for the spin susceptibility of Ni, Pd, and Pt are similar to those obtained by Mori.<sup>18</sup>

We have also calculated the real part of the dynamical susceptibility for finite values of  $\omega$  as a function of momentum transfer and temperature. As in the preceding calculations the qualitative behavior has been found to be the same for all the metals, therefore we present here the results only for Pd for  $\omega = 0.03$  eV (Fig. 6). With the increase of  $\vec{q}$  and temperature the magnitude of  $\operatorname{Re}\overline{\chi}^0(\vec{q},\omega,T)$  decreases and the peak at low  $\overline{q}$  and T becomes broadened. Re $\overline{\chi}^{\nu}(\vec{q}, \omega, T)$  at higher momentum transfer is independent of temperature. For small values of  $\vec{q}$ ,  $\text{Re} \vec{\chi}^{0}(\vec{q}, \omega, T)$  decreases almost linearly with the increase of temperature. This behavior is again qualitatively consistent with the calculations of Hirooka and Shimizu.<sup>24</sup> The band-structure calculation whose effect is predominant at low temperatures enhances the magnitude of  $\operatorname{Re}\overline{\chi}^0(\vec{q},\omega,T)$  at low temperatures.



FIG. 6. Variation of the real part of dynamical reduced spin susceptibility  $\text{Re}\overline{x}^{0}(\vec{q}, \omega, T)$  with  $\vec{q}$  and  $T/T_{F_{i\sigma}}$  for particular energy  $\omega = 0.03$  eV for Pd. In (a) curves labeled 1, 2, 3, 4 correspond, respectively, to  $T/T_{F_{i\sigma}} = 0.2, 0.4, 0.8, 1.2$  and in (b) the curves labeled 1, 2, 3, 4 correspond, respectively, to q = 0.05, 0.25, 0.85, 1.25.

# A. Exchange-enhanced suscpetibility function

The exchange-enhanced susceptibility is given as

$$\chi(\vec{\mathbf{q}},\omega,T) = \frac{\chi^0(\vec{\mathbf{q}},\omega,T)}{1 - I\chi^0(\vec{\mathbf{q}},\omega,T)} \quad , \tag{25}$$

where I is the exchange interaction parameter which depends upon temperature. Since the explicit form of the temperature dependence of I is not known fully therefore in the present calculations we treat I to be temperature independent. The Stoner enhancement factor for s and d electrons is defined as

$$S_{s} = \frac{\chi_{s}(0,0,0)}{\chi_{v}^{0}(0,0,0)} = \frac{1}{1 - I\chi_{v}^{0}(0,0,0)} \quad , \tag{26}$$

$$S_{dm} = \frac{\chi_{dm}(0,0,0)}{\chi_{dm}^{0}(0,0,0)} = \frac{1}{1 - I\chi_{dm}^{0}(0,0,0)} \quad . \tag{27}$$

Recently Janak<sup>25</sup> calculated *I* for a number of 3*d*- and 4*d*-series transition metals and we adopt the same values in the present calculations. Since the results for Ni, Pd, and Pt have similar qualitative features therefore we present the results for the exchange-enhanced susceptibility for Pd only, which are shown in Fig. 7. The separate exchange correlation strengths are used for the *s*- and *d*-band contributions. The Stoner enhancement factor for *s* band is  $S_s = 1.0368$  and for the *d* subbands is  $S_{dm} = 1.5398$ . The general behavior of the exchange enhanced susceptibility function is the same as that of unenhanced



FIG. 7. Exchange enhanced static susceptibility  $\overline{\chi}(\vec{q}, 0, T)$  as a function of temperature for Pd. The curves labeled 1, 2, 3, 4 correspond, respectively, to q = 0.05, 0.25, 0.65, 1.05.

susceptibility function. The magnitude decreases with  $\vec{q}$  and T. The peaks at low  $\vec{q}$  and T get more pronounced in the exchange enhanced susceptibility. However with the increase of  $\vec{q}$ , the peaks get broadened. This emphasizes the importance of exchange correlation corrections in the response function of the magnetic metals.

# **IV. DISCUSSION**

Vosko and Perdew<sup>26</sup> suggested that

$$\chi(0,0,0) = \frac{N(E^{F}(0))}{1 - IN(E^{F}(0))}$$
(28)

is a lower bound to the susceptibility. Any material for which  $IN(E^{F}(0)) > 1$  should be ferromagnetic in nature where  $N(E^{F}(0))$  is the density of states at the Fermi surface. In the model band scheme for ferromagnetic phase of Ni,  $IN(E^{F}(0))$  comes out to be equal to 1.01, which shows the existence of ferromagnetism. For paramagnetic Ni and Pd the values of  $IN(E^{F}(0))$  are 0.55 and 0.35, respectively. However,  $IN(E^{F}(0))$  could not be evaluated for Pt, as a careful estimate of the exchange parameter I is not available. It is shown by Janak<sup>25</sup> that ferromagnetism occurs basically because of the spatial localization of the d orbitals near the top of the d band. This localization produces both the large density of states (when the band is nearly full) and a relative maximum in the exchange correlation parameter I. The product  $IN(E^{F}(0))$  is large enough for ferromagnetism only at the end of 3d series but not the 4d and 5d series. This is because the 4d and 5d wave functions have spatial distribution to a larger extent which implies larger interaction between the neighbors, a larger bandwidth and a smaller density of states. In the band model scheme of ferromagnetic Ni, the effective masses of the d subbands are large which means d subbands are flat and thereby the d-band width in ferromagnetic Ni is smaller compared to that in paramagnetic Ni, Pd, and Pt. The large value of spin susceptibility for ferromagnetic Ni (large density

of state at the Fermi surface) and small d-band width in our model favors the viewpoint of Janak.<sup>25</sup> This gives confidence in the model band scheme which we have used in the present investigations. In studying the temperature-dependent susceptibility, we have neglected completely the effect of s-d hybridization. The *d* band is represented parabolically and the explicit temperature variation of spin splitting for ferromagnetic Ni is ignored. In the exchange enhancement of  $\chi(\vec{q}, 0, T)$ , which is introduced here phenomenologically, the Coulomb interaction parameter is assumed to be temperature independent. Actually the parameter I is sensitive to temperature and its value decreases as the temperature increases. These approximations may further be improved to obtain better qualitative results. However, these approximations greatly simplify the numerical computations and the effect of d electrons could be studied explicitly at least qualitatively. To perform a selfconsistent calculation for a transition metal, one should also include the temperature dependence of band structure and many-body effects. However to take all of these features into account rigorously would be a difficult problem in itself.

Our calculations may resemble the one band model calculation but our results certainly include the sound physical bases by including both the s- and d-electron contributions. In the one band model one of these contributions is neglected. Our calculated  $\chi(\vec{q}, 0, T)$  in the  $q \rightarrow 0$  limit for Pd and Pt explains the qualitative features obtained in the experimental measurements of Van Dam<sup>27</sup> (i.e., a sharp peak in Pd and a diffused peak in Pt at low temperature).

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