High-energy paramagnetic spin fluctuations in nickel

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The spin fluctuations with large wave vectors in paramagnetic nickel above T_c have been studied by means of computer calculations of the dynamical susceptibility. The spectral function and the cross section for neutron scattering have well-defined peaks that indicate the special character of the dynamics of $3d$ electrons in Ni. The peak has not, however, resonance shape. The structure of spin excitations is determined by critical spin dynamics as well as by electronic energy bands.
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I. INTRODUCTION

The nature of the spin fluctuations in the paramagnetic phase of the ferromagnetic $3d$ metals is still not satisfactorily understood. The reason for this is the nonlocalized magnetic moment and the special role of the kinematics of the electrons (the $3d$ band is very narrow compared to the s band). A particular focus of attention has been on the possibility of the persistence of spin waves into the paramagnetic phase.[1] Nickel, having completely delocalized magnetic moments, is an appropriate sample for experimental investigation of the specific character of the spin dynamics of $3d$ metals. The detection of a well-defined peak with a quadratic dispersion law in the cross section for the scattering of neutrons in the paramagnetic phase of Ni in the constant energy scan[2] has suggested the possibility of the existence of spin waves above the Curie temperature. In a constant q vector scan, however, the experimental results, for moderate q ($0.4<q<0.6$ Å^{-1}) have been controversial. There are measurements in which the cross section shows a peak at nonzero energy; these have been interpreted in terms of propagating spin excitations,[3] but their existence was not confirmed by other measurements.[4] Because of this controversy, computer calculations of the cross section, based on realistic energy bands and the real crystal structure of Ni, have been carried out.[5] For small q vector $q\leqslant 0.3$ Å^{-1}, the computer cross section agreed well with the experimental one. For moderate q, the position of the peak in the cross section is in accord with the experimental results; however, this peak appeared to be too wide to be related to the propagating spin excitations. Since then, new measurements of the cross section have been done for large q vectors ($q\approx 0.78$ Å^{-1}),[6] and the results have been treated as convincing evidence for the existence of propagating spin-wave excitations in the paramagnetic phase of Ni. Because the results of these measurements are regarded as crucial to the understanding of the nature of the spin fluctuations in the $3d$ metals in the paramagnetic phase, it would be desirable to have a microscopic theory that covers a wide range of q vectors.

The problem is complex since it concerns the *time-dependent, large* spin fluctuations in the high-temperature (paramagnetic) phase of Ni. At this temperature, the criticality of the fluctuations is important (or at least it is not negligible). Consequently, many-body effects should be taken into account (to obtain the correct thermodynamics). The most reliable dynamical mean-field approximation, which is based on density-functional theory, gives a satisfactory description of the static properties of paramagnetic Ni (under additional assumptions concerning the structure of paramagnetic state).[24] A correct (i.e., ω is not considered as a parameter only) extension of this theory to time-dependent quantities requires the use of time-dependent density-functional theory.[25] For our case, its application is a problem that has not been solved yet.

We follow a many-body approach.[1] This approach, which focuses mostly on the correct treatment of time-dependent spin fluctuations (their correlations) rather than on the self-consistency of the energy bands and spin fluctuations, yields the correct thermodynamics, however. Our aim is the analysis of time-dependent spin fluctuations with large q vector, $q\gtrsim 0.78$ Å^{-1}, by means of computer calculations of the dynamical susceptibility. We have computed the cross section for the inelastic magnetic scattering of unpolarized neutrons in paramagnetic Ni at a temperature of 700 K in the (1,1,1) direction (in constant q vector scan). The calculations are based on a dynamical mean-field-like approximation with the q- and ω-dependent effective interaction evaluated in the polarization potential approximation[7] (for $T\neq 0$). The two q-dependent parameters of the effective interaction are evaluated using results of the renormalization-group theory.[8,9] In our calculations we are able to describe time-dependent spin fluctuations taking into account their correlations and criticality as well as the energy-band effects.

II. BACKGROUND THEORY

The fluctuations of the magnetic-moment density are described by the correlation function of magnetization (the structure factor), which is connected, via the fluctuation-dissipation theorem, to the imaginary part of the dynamical susceptibility $\chi(\mathbf{q},\omega)$. The quantity that is measured directly in inelastic-neutron-scattering experiments is the structure

function $S(\mathbf{q},\omega)$ that is related to the dynamical susceptibility

$$S(\mathbf{q},\omega)=\frac{2}{1-e^{-\beta\omega}}\ \mathrm{Im}\ \chi(\mathbf{q},\omega). \tag{1}$$

It is useful to represent $S(\mathbf{q},\omega)$by the shape function $\phi(\mathbf{q},\omega)$

$$S(\mathbf{q},\omega)=2\chi(\mathbf{q})\phi(\mathbf{q},\omega)\frac{\omega}{1-e^{-\beta\omega}}. \tag{2}$$

The dynamical susceptibility $\chi^{+-}(\mathbf{q},\omega)$ can be expressed by the formally *exact* expression[1]

$$\chi^{+-}(\mathbf{q},\omega)=\frac{\chi_0^{+-}(\mathbf{q},\omega)}{1-I(\mathbf{q},\omega)\chi_0^{+-}(\mathbf{q},\omega)}. \tag{3}$$

The functional $I(\mathbf{q},\omega)$describes the correlations between electrons and $\chi_0^{+-}(\mathbf{q},\omega)$ is the dynamical susceptibility of noninteracting electrons. For strongly correlated electron systems the evaluation of $I(\mathbf{q},\omega)$ is the main problem in the calculation of a dynamical susceptibility. In the Hubbard model, in the random phase approximation, $I(\mathbf{q},\omega)$ is q and ω independent; i.e., $I(\mathbf{q},\omega)\simeq I_0=\mathrm{const}$. Even in that approximation the spin dynamics of $3d$ metals, in the ferromagnetic phase, is described sufficiently well near the spin-wave resonances. In the paramagnetic phase, however, one has to know the spin dynamics over a wide range of energy ω. It requires essential renormalization of I_0 making it q and ω dependent.

It has been shown that the neutron-scattering cross section calculated with $\chi^{+-}(\mathbf{q},\omega)$, given by Eq. (3), fits the measured cross section of paramagnetic Ni well[5] if $I(\mathbf{q},\omega)$ is assumed to be independent of ω. Thus $I(\mathbf{q},\omega)$ is sensitive to the value of q, but it is weakly dependent on ω as long as q is not too large ($q\leqslant 0.5$ Å^{-1}). Such behavior of $I(\mathbf{q},\omega)$ is expected because $I(\mathbf{q},\omega)$ depends on higher correlation functions. It allows us to use the approximation $I(\mathbf{q},\omega)\cong I(\mathbf{q},\omega_c(q))=I(q)$ (where ω_c is the characteristic energy of the system).

Now the static susceptibility defined as $\chi(\mathbf{q})=\int\chi(\mathbf{q},\omega)d\omega$ is related to $I(q)$ through Eq. (3) with $I(q,\omega)\simeq I(q)$. This relation gives the possibility of obtaining $I(q)$ from the measured $\chi(q)$and hence of calculating the *dynamical* susceptibility $\chi(q,\omega)$ on the basis of the *static* one. Although this procedure was successfully used for $q<0.6$ Å^{-1},[5] it does not give satisfactory results for large q ($q>0.6$ Å^{-1}). There are three reasons for this: (a) the dependence of $I(q,\omega)$ on ω is not negligible, (b) there are no reliable measurements of $\chi(q)$ for large q,[10] (c) $\int\chi(q,\omega)d\omega$, unlike Im $\chi(q,\omega)$, is not sufficiently sensitive to the form of $I(q)$.

III. POLARIZATION POTENTIAL APPROXIMATION FOR $I(q,\omega)$

A model for the q and ω dependence of $I(q,\omega)$, for small q,ω (q/q_F, $\omega/\omega_F\ll 1$) is given by the Landau theory of Fermi liquid[11]

$$I(q,\omega)=F_0+F_1(\omega/q)^2 \tag{4}$$

(for the relation between paramagnon and Fermi-liquid theory, see Ref. 11). F_0 and $F_1$are the Landau amplitudes of the antisymmetric part of the effective interaction between quasiparticles with $l=0,1$. Unfortunately for large q ($q>0.6$ Å^{-1}; i.e., q more than halfway to the boundary of the first Brillouin zone), the Landau model $I(q,\omega)$, Eq. (4), does not give satisfactory results, as can be checked by computer calculations.

A simple extension of the Landau theory for large q,ω is the polarization potential theory[7] that successfully describes interacting fermions systems like He^3 (Ref. 7) or electrons in paramagnetic metals.[12] It takes into account more complicated processes in the scattering of particle-hole pairs than their multiscattering by the direct potential (the Landau approximation) and it includes also multipair processes. Note, however that polarization potential theory is a mean-field theory.

The polarization potential theory assumes that the induced scalar potential $\phi_{\mathrm{pol}}(q,\omega)$ and induced vector potential $\mathbf{A}_{\mathrm{pol}}(q,\omega)$are proportional to induced charge and current densities $\langle\rho(q,\omega)\rangle$, $\langle\mathbf{j}(q,\omega)\rangle$

$$\begin{aligned}\phi_{\mathrm{pol}}(q,\omega)&=F_0(q)\langle\rho(q,\omega)\rangle,\\ \mathbf{A}_{\mathrm{pol}}(q,\omega)&=F_1(q)\langle\mathbf{j}(q,\omega)\rangle,\end{aligned} \tag{5}$$

which in turn couple to the charge density ρ_q and to the current density $\mathbf{j}_q$ via terms $\rho_q\phi_{\mathrm{pol}}(q,\omega)$and $\mathbf{j}_q\mathbf{A}_{\mathrm{pol}}(q,\omega)$. Note that one is assuming that $F_0(q)$ and $F_1(q)$do not depend on ω. The conservation law of charge couples $\langle\rho(q,\omega)\rangle$ to $\mathbf{j}(q,\omega)$leading to the response function given by Eq. (3) with $I(q,\omega)$ written as

$$I(q,\omega)=F_0(q)+F_1(q)(\omega/q)^2, \tag{6}$$

if Eqs. (5) are considered for the electron spins and $\chi_0^{+-}(q,\omega)$ is replaced by the response of a system to the local (screened) field. Hence, $I(q,\omega)$in the polarization potential approximation has the same form as the Landau approximation, Eq. (4), but F_0, F_1 are now renormalized in such a way that they become q dependent. The polarization potential theory can be justified by a microscopic treatment of strongly correlated fermion systems[13] where, by solving the Bethe-Salpeter equation for the vertex function for large q,ω, one obtains $I(q,\omega)$ in the form of Eq. (6). The form of the functions $F_0(q),F_1(q)$are strongly restricted by the exchange symmetry of a system. The evaluation of $I(q,\omega)$ in the framework of microscopic theory will be done by us elsewhere.

Although the Landau and polarization potential theories were developed for zero temperature, the form of $I(q,\omega)$ for $T\neq 0$ can be taken as that for zero temperature, but now the functions $F_0(q),F_1(q)$are temperature dependent. This is because the form of $I(q,\omega)$ follows from the relations between the potentials of electromagnetic field, the induced densities (of charge and spins) and the currents, and those relations are very general. The screened susceptibility $\chi_{sc}^{+-}(q,\omega)$ contains contributions from multiparticle processes. Since the amplitude of multiparticle processes that are relevant for the susceptibility are proportional to the small parameter $(K_BT\rho_F)^{4/3}\sim 0.05$($T=700$ K, ρ_F is the density of states at Fermi surface)[14] we have used the approxi-

mation $\chi_{sc}^{+-}(q,\omega)\sim\chi_0^{+-}(q,\omega)$, which seems to be sufficiently good in our case.[7] In this sense the polarization potential theory of the dynamical susceptibility is an approximation for the general expression Eq. (3), giving a model for the function $I(q,\omega)$. In our calculations the temperature-dependent function $I(q,\omega)$ in Eq. (4) has been used in the polarization potential approximation, Eq. (5). It is clear that this approximation goes beyond paramagnon theories.

IV. EVALUATION OF THEORY PARAMETERS $F_0(q),F_1(q)$

Over the range of parameters T,q,ω in which we are interested, the correlation length ξ is not small and $\xi_q\sim 6$ for $q=0.78$ Å^{-1} in nickel. The scaling hypothesis works in dynamics for $\xi_q\gg 1$ and so the system is still in the region where the criticality of spin fluctuations plays an important role. To determine the functions $F_0(q)$ and $F_1(q)$in $I(q,\omega)$, a phenomenological model of the critical spin dynamics of a ferromagnet in the asymptotic renormalization-group approximation[8,9] has been used. The cross section for neutron scattering, calculated in the framework of this theory, describes sufficiently well the measured cross section in the paramagnetic phase of Ni for small to intermediate values of q ($q<0.6$ Å^{-1}).[4,5,9] According to the phenomenological theory of spin fluctuations in the paramagnetic phase of a ferromagnet (in the critical region), the shape function $\phi(x,s)$ for $T>T_c$ and *arbitrary* q,ω has the form[8,9]

$$\phi(x,s)=\mathrm{Re}\,\frac{1}{-is+[Z(x)\Pi(x,isZ(1+x^{-2})]^{-1}}, \qquad (7)$$

where

$$\Pi(x,iw)=[(1+bx^{-2})^{5/4}-aiw]^{3/5},$$

and $x=q\xi$, $s=\omega/\omega_c$. Here ξ is the correlation length, which scales according to $\xi=1.49(T/T_c-1)^{-0.735}$ for Ni, and ω_c is the characteristic frequency $\omega_c=Aq^{2.5}\Omega(q\xi)$. The scaling function $\Omega(x)$ is obtained by the requirement that ω_c is the half width of the structure factor

$$\Omega(x)=(1+x^{-2})Z(x)/Z(\infty),$$

where

$$Z(x)=\left[1-\alpha\ \arctan\left(a\,\frac{1+x^{-2}}{(1+bx^{-2})^2}\right)\right]^{-1}(1+bx^{-2})^{-3/4}.$$

The a,α,b, being the model parameters, depend very weakly on x, however. Their values are $a=0.46$, $\alpha=0.51$, $b=3.16$. The parameter A is the free, adjustable parameter that can be fixed by a fit to experiment. At low to intermediate q ($q<0.6$ Å^{-1}) the best fit is $A=390$ meV Å^{-1}.

The effective interaction parameters F_0,F_1 usually are calculated in a microscopic model or else they are determined by the fit of a measured quantity to the experimental value. In our case $I(q,\omega)$, and consequently $F_0(q),F_1(q)$, has been determined *analytically* by comparing the spectral function, calculated from Eq. (3), with the spectral function, Eq. (7), obtained in the renormalization-group model

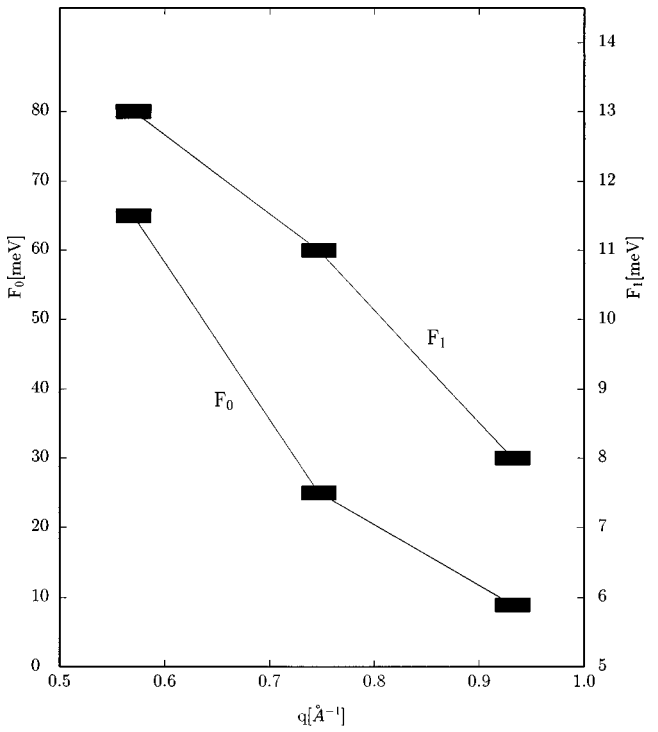

FIG. 1. The q dependence of the polarization potential parameters $F_0(q),F_1(q)$; see Eq. (6).

$$\mathrm{Im}\ \chi(q,\omega)/\omega=\chi(q)\,\phi(q\xi,\omega/\omega_c). \qquad (8)$$

We can expect that an effective interaction is a smoother function of q and ω than, for instance, the dynamical susceptibility, which makes it more useful in an estimation procedure. Im $\chi(q,\omega)$in Eq. (8) was evaluated using Eq. (3) where an (ω/q) expansion for Im $\chi_0(q,\omega)$, Re $\chi_0(q,\omega)$ was used with coefficients determined by the energy-band structure.[1] $\chi(q)$, at high q, was approximated by an Ornstein-Zernike extrapolation of the low-q experimental data (but with a larger correlation length than that obtained by scaling, as is suggested by experiment[10]). The procedure described above for the calculation of an effective interaction can be used also in the high-q regime ($q\geqslant 0.6$ Å^{-1}), which is the focus of this paper. We have to keep in mind, however, that the renormalization-group theory works better for small x, s. $I(q,\omega)$ obtained in that way is a decreasing function of q (''screening effect'') for $q>0.5$ Å^{-1} (see Fig. 1). There is analogous q dependence of the function $I(q,\omega)$ in strongly interacting fermions systems like He^3 (Ref. 7) or paramagnetic metals.[12] Having the effective interaction $I(q,\omega)$, the dynamical susceptibility $\chi(q,\omega)$ [and the cross section $S(q,\omega)$] was calculated using Eq. (3), where the computed $\chi_0^{+-}(q,\omega)$ given by Eq. (9) has been used. The Im $\chi(q,\omega)$ and $S(q,\omega)$ calculated in that way are stable with respect to small variations of A and $\chi(q)$ and are in good agreement with experiment for $q<0.6$ Å^{-1}.[5]

In the polarization potential approximation the contribution of interaction processes to Im $\chi(q,\omega)$, which renormalize the effective interaction, are neglected. This approximation seems to be reasonable because the amplitudes of multiparticle processes relevant for $\chi^{+-}(q,\omega)$ are proportional to the power of the small parameter $(K_BT\rho_F)^{4/3}$

~ 0.05. It means that the cross section calculated with $\chi^{+-}(q,\omega)$ from Eq. (1), where $I(q,\omega)$ is used, has an underestimated half width.

The off-diagonal elements of $\chi^{+-}_{0\,st}(q,\omega)$ yield an additional contribution to the effective field acting on the spin (local-field correction).[15] It is strictly a crystal effect and it is important for high temperature and large q. Consequently, $\chi^{+-}_{0\,st}(q,\omega)$ is calculated beyond the form-factor approximation.[16]

The question whether or not the magnetic splitting Δ in the paramagnetic phase of Ni is zero, at least locally, is still not satisfactorily solved.[17] Since no splitting has been found to date in the paramagnetic phase of Ni, in contrast to the case of Fe,[18] we have put the magnetic splitting equal to zero in our calculations.

V. RESULTS AND DISCUSSION

We now proceed to calculate the scattering function $S(q,\omega)$ and the spectral function Im $\chi^{+-}(q,\omega)$ using Eqs. (1) and (3), the model functions $F_0(q)$ and $F_1(q)$ displayed in Fig. 1, and realistic electronic energy bands. The susceptibility of noninteracting electrons

$$\chi^{+-}_{0\,st}(\mathbf{q},\omega) = -N^{-1}\sum_{l,m,\mathbf{k}} \frac{n_m(\mathbf{k}) - n_l(\mathbf{k}+\mathbf{q})}{\epsilon_m(\mathbf{k}) - \epsilon_l(\mathbf{k}+\mathbf{q}) + \psi + i\eta} \times \langle m,\mathbf{k}|e^{-i\mathbf{q}_s\mathbf{r}}|l,\mathbf{q}+\mathbf{k}\rangle\langle l,\mathbf{k}+\mathbf{q}|e^{i\mathbf{q}_t\mathbf{r}}|m,\mathbf{k}\rangle, \tag{9}$$

is calculated using semiempirical energy bands $\epsilon(\mathbf{k})$.[19] These bands are in good agreement with the photoemission experiment.[20] In Eq. (9) we have taken into account the umklapp processes: $\mathbf{q}=\mathbf{q}+\mathbf{K}$, where $\mathbf{K}$ is a reciprocal lattice vector.

We have carried out the calculation of the scattering function $S(q,\omega)$ and the spectral function Im $\chi^{+-}(q,\omega)$ using $\chi^{+-}(q,\omega)$ from Eq. (3), and $I(q,\omega)$ calculated from Eq. (8). The results of our calculations for $T=700$ K and $q=0.772$ Å^{-1} are shown in Figs. 2 and 3. There is a well-defined peak at $\omega_0 \sim 140$ meV in the scattering and spectral functions in good agreement with the $\omega_0 \sim 135$ meV suggested by experimental measurements.[6] It indicates that despite the specific electronic structure of Ni, the spin fluctuations with energies near ω_0 dominate according to macroscopic theory.[9] Nevertheless, the specific kinematics of the electrons in the $3d$ energy bands in Ni and the correlations between them influence much of the dynamics of the spin fluctuations. This influence is expected to be significant for $q > 0.3$ Å^{-1} because with increasing q vector, in that region, the enhancement of $\chi^{+-}_{0\,st}(q,\omega)$ decreases whereas the peak position is shifted to higher energies. The essential reduction of the half width of the peak, compared to the results of phenomenological theory, is due to band effects. It indicates that $3d$ electrons in Ni yield a considerably less effective dissipation channel in spin systems than phenomenological theory has assumed. Note that the half width of the spectral function calculated with parabolic energy bands is very large compared to that obtained with realistic $3d$ bands. The energy-band structure is responsible also for the significant decrease in the value of Im χ^{+-} and $S(q,\omega)$ at

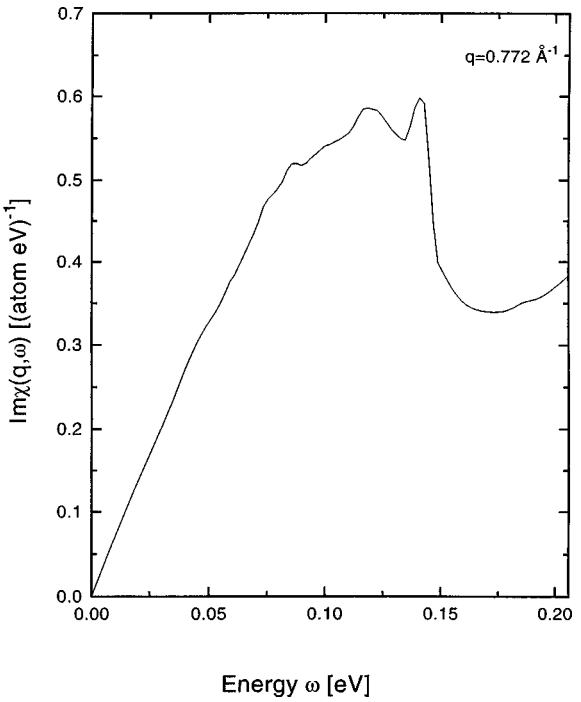

FIG. 2. The calculated imaginary part of the dynamical susceptibility; $q=0.772$ Å^{-1}.

$\omega_1 = 140$ meV that substantially influences the shape and thus also the position of the peak. This is caused by a significant decrease in the value of the dynamical susceptibility of noninteracting electrons $\chi^{+-}_{0\,st}(q,\omega)$, in this ω range, since Im $\chi^{+-}_{0\,st}(q,\omega)$ behaves analogously to Im $\chi^{+-}(q,\omega)$ in the neighborhood of $\omega \sim 140$ meV. In our case, unlike the experimental results,[6] the low-energy ω ($\omega < 100$ meV) spin fluctuations can be relatively easily excited (they have relatively

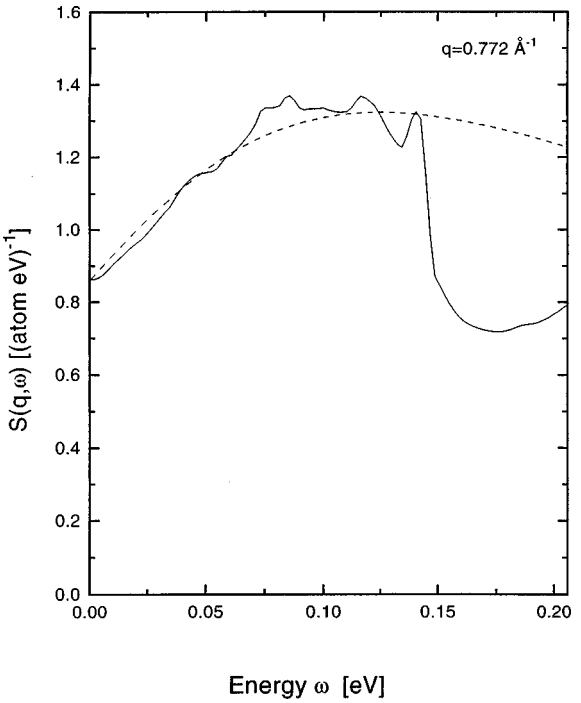

FIG. 3. The scattering function computed using realistic energy bands (solid line) and calculated in the framework of renormalization group (dashed curve); $q=0.772$ Å^{-1}.

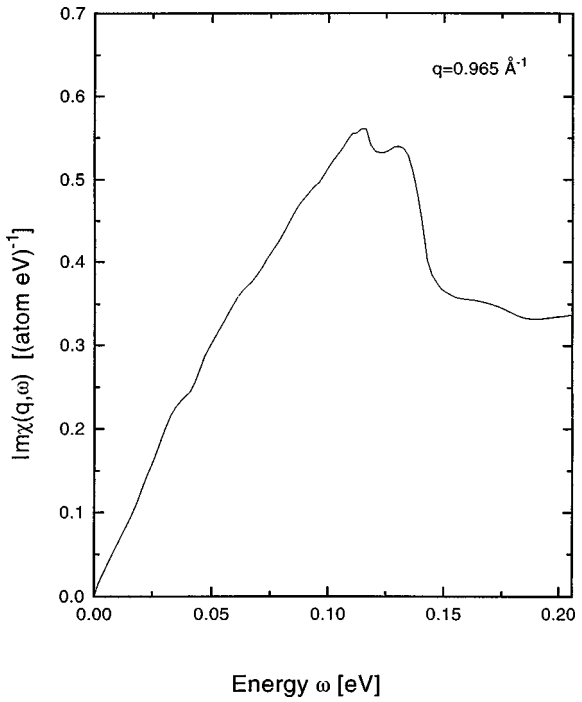

FIG. 4. The calculated imaginary part of the dynamical susceptibility for large q vector; $q=0.965$ Å^{-1}.

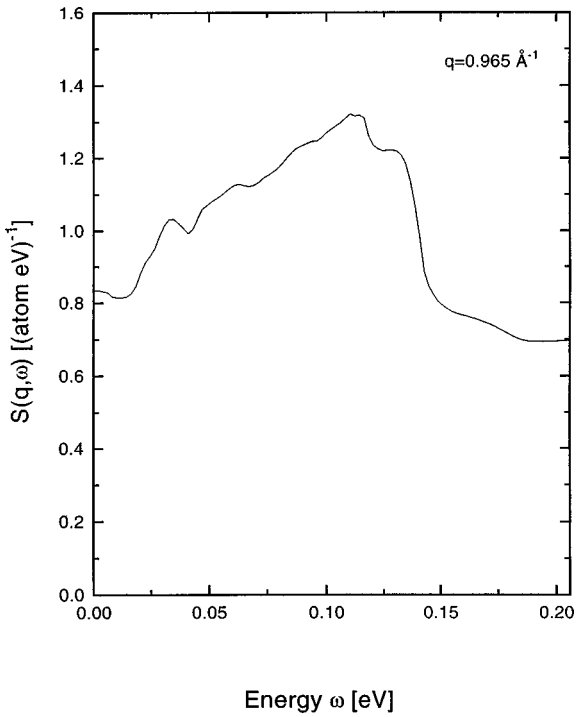

FIG. 5. Computed scattering function of large q vector; $q=0.965$ Å^{-1}.

high spectral weight). It results in the wide peak of the spectral function Im $\chi^{+-}(q,\omega)$. This peak has considerably smaller half width than that predicted by macroscopic theory; it has the half width Γ ($\Gamma\sim 140$ meV) which is comparable with its position $\omega_0\sim 140$ meV. Hence $\Gamma\geqslant\omega_0$ and therefore it is difficult to interpret it in terms of propagating spin excitations. The calculated Im $\chi^{+-}(q,\omega)$ and $S(q,\omega)$ for $q\simeq 0.78$ Å^{-1} are stable with respect to changes in the adjustable parameter A and are consistent with our results for $\chi^{+-}(q,\omega)$, in the region of not large q ($q<0.6$ Å^{-1}).

To make our results more reliable, we have calculated Im $\chi^{+-}(q,\omega)$ and $S(q,\omega)$ for very large q vector $q=0.965$ Å^{-1} (Figs. 4 and 5). For this q vector the peak in Im $\chi^{+-}(q,\omega)$ is more pronounced in comparison to that for $q=0.772$ Å^{-1} and its position is shifted slightly to larger ω, mainly due to the change of its shape. These results agree with experiment.[6] However, the increase in the spectral weight of spin fluctuations with large ω ($\omega>150$ meV) when q is increased, is small compared to those suggested by experiment. It is for this reason that the shift of the peak position is small compared to experimental results, and consequently its half width is still comparable with its position. Such behavior of the spectral function is caused by the $3d$ electron kinematics in Ni; its peak shifts to higher energy more strongly for parabolic energy bands than for realistic ones (but giving in that case a very wide peak). Note that the structure of the spectral function of the spin fluctuations indicates that the contribution of high-energy spin fluctuations to the integral quantity $\chi(q)$ and $S(q)$ is significant. In particular, integration of $\chi(q,\omega)$ up to a value of 100 meV gives an underestimated value of $\chi(q)$.[10]

VI. CONCLUSIONS

In conclusion, we have studied spin dynamics for large q vector in paramagnetic Ni in a mean-field-like approximation. The effective interaction between electrons was evaluated in the framework of polarization potential theory, which is relevant for the description of correlated electrons (and it goes beyond random phase approximation). The unknown effective interaction parameters $F_0(q),F_1(q)$ were determined by the requirement that our theory reproduces the renormalization-group theory results which, in turn, describe experiments satisfactorily (at least for not large q). Our calculations show the existence of a peak in the cross section for neutron scattering and yield its position in good agreement with experiment. The appearance of that peak originates from the interplay between two effects. The analysis of critical spin dynamics shows that the spin fluctuations with energy ω_0 should dominate as the consequence of the conservation law of magnetic moment.[9] On the other hand, the dynamics of these fluctuations should obey the strong constraints imposed on it by the kinematics of the $3d$ electrons in Ni. It results in a considerably better defined peak in the cross section than that predicted by macroscopic theory.[9] However, it is too wide for interpretation in terms of propagating spin excitations.

In our calculations the exchange splitting Δ was put equal to zero for paramagnetic Ni, according to experiment. An improvement to our results can be expected by taking into account the existence of short-range order above T_c.[21,22] Our considerations suggest that a new, more basic theory of spin fluctuations above T_c in $3d$ metals is needed, and in particular one that treats self-consistently the dynamics of electrons and spins, especially in the critical region.[23]

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