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# Spin and orbital contributions to the dynamical structure factors of paramagnetic transition metals

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Calculations have been made of the wave-vector-dependent dynamical structure factor for the itinerant transition metals V, Nb, Rh, and Pd. For inelastic neutron scattering there is a spin and an orbital contribution. The purpose of this work is a comparison of the relative magnitude of the two contributions. An important conclusion from the numerical work is the likely importance of the orbital part for a wide range of energy and momentum transfers.

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## I. INTRODUCTION

The wave-vector-dependent dynamic spin susceptibility is a quantity that is sensitive to the detailed physical characteristics of an electron gas. The calculation of the susceptibility of transition metals based on realistic band structures is a formidable task and it is only over the last decade that schemes have been developed for its accurate evaluation. Stenzel and Winter[1] present the results of calculations for Pd and V and give references to earlier work. They use a local-spin-density functional theory together with the random-phase approximation to give a description of the enhancement effects within the inhomogeneous electron gas. A comparison of different formulations of the theory has since been given by Winter *et al.*[2] Calculations of the spin-flip susceptibility for the cubic ferromagnets iron and nickel have been performed by Cooke *et al.*[3-5]

The most direct probe of the spin susceptibility is inelastic neutron scattering. Neutrons, however, interact with both the electron spin and the electron current, so that the total susceptibility comprises both a spin and a so-called orbital contribution. The orbital part is often ignored, although a few calculations do exist that attempt its evaluation. Cooke *et al.*[6] have studied the static susceptibility for paramagnets and Morgan *et al.*[7] and Blackman *et al.*[8] have considered the dynamic susceptibility of semiconductors and simple metals, respectively. An effective-mass argument has been used in the past to justify ignoring the orbital contribution in comparison with the spin part when studying *d*-band metals. A calculation by Blackman *et al.*[9] using the paramagnetic band structure of iron indicates that this assumption is not tenable.

For free electrons the orbital part of the dynamic susceptibility dominates at low wave-vector transfer, Q, but at higher Q it is the spin part that is dominant. This behavior arises because of a $Q^{-2}$ factor in the orbital part.[8] In the work by Blackman *et al.*,[9] there were indications that this simple trend no longer applies in the transition metals. Indeed, the possibility exists that the orbital contribution can dominate over most of the range of the wave-vector transfer of interest.

In the present paper we extend our calculations to the paramagnetic transition metals V, Nb, Rh, and Pd, and show that the orbital contribution is seldom negligible and is often dominant. At this stage we consider just the unenhanced susceptibility. Obviously it will be important eventually to include enhancement effects, but the present calculations, taken together with information[1] about enhancement to the spin susceptibility will give a fairly clear picture of the relative importance of the orbital susceptibility for the transition metals.

## II. THEORY AND CALCULATION

The full theory on which the calculations are based was described earlier.[9] We reproduce here just sufficient details to make this paper self-contained. The partial differential magnetic cross section for the scattering of unpolarized neutrons from a system containing $N$ electrons is given by

$$\frac{d^2\sigma}{d\Omega dE}=Nr_0^2(k_f/k_i)S(\mathbf{Q},\omega) \qquad (1)$$

where $\mathbf{Q}=\mathbf{k}_f-\mathbf{k}_i$ is the wave-vector transfer and $r_0^2=0.29$ b. The dynamic structure factor $S(\mathbf{Q},\omega)$ is written as the sum of a spin and an orbital contribution.

The electronic Bloch functions are expanded in terms of symmetry orbitals within the Wigner-Seitz sphere

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$$\Psi_{n\mathbf{k}}(\mathbf{r})=\sum_{\mu} i^{l(\mu)}a_{n\mu}(\mathbf{k})R_{l(\mu)}(r)Y_{\mu}(\hat{\mathbf{r}}) \ , \tag{2}$$

where $\mu$ denotes the pair of angular momentum labels $l,m$ and $l(\mu)$ indicates the value of $l$ associated with $\mu$. The factors $R$ are radial functions and $Y$ are spherical harmonics. Using the expansion coefficients $a_{n\mu}(\mathbf{k})$, we can define a symmetry-orbital-dependent single-particle Green's function $\Gamma$,

$$\Gamma_{\mu\nu\eta\xi}(\mathbf{q},\omega)=\frac{1}{N}\sum_{\substack{n,p\\ \mathbf{k}}}\frac{a_{n\mu}(\mathbf{k})a_{p\nu}(\mathbf{k}+\mathbf{q})a_{n\eta}(\mathbf{k})a_{p\xi}(\mathbf{k}+\mathbf{q})}{\hbar\omega-E_{p\mathbf{k}+\mathbf{q}}+E_{n\mathbf{k}}} \times(f_{n\mathbf{k}}-f_{p\mathbf{k}+\mathbf{q}}) \ , \tag{3}$$

where $E_{n\mathbf{k}},f_{n\mathbf{k}}$ are band energies and Fermi occupation numbers associated with band $n$ and wave-vector $\mathbf{k}$.

The spin and orbital components of $S(\mathbf{Q},\omega)$ may then be written

$$S^{(s)}(\mathbf{Q},\omega)=-\frac{1}{\pi}\sum_{\mu,\nu,\eta,\xi}\mathrm{Im}\Gamma_{\mu\nu\eta\xi}(\mathbf{q},\omega)F^{(s)}_{\mu\nu}(\mathbf{Q})F^{(s)}_{\eta\xi}(\mathbf{Q}) \ , \tag{4}$$

$$S^{(o)}(\mathbf{Q},\omega)=-\frac{1}{\pi\hbar Q^2}\sum_{\mu,\nu,\eta,\xi}\mathrm{Im}\Gamma_{\mu\nu\eta\xi}(\mathbf{q},\omega)\{[\mathbf{F}^{(o)}_{\mu\nu}(\mathbf{Q})]^{\dagger}\cdot\mathbf{F}^{(o)}_{\eta\xi}(\mathbf{Q})-[\hat{\mathbf{Q}}\cdot\mathbf{F}^{(o)}_{\mu\nu}(\mathbf{Q})]^{\dagger}[\hat{\mathbf{Q}}\cdot\mathbf{F}^{(o)}_{\eta\xi}(\mathbf{Q})]\} \ . \tag{5}$$

In these equations $\mathbf{q}$ is a wave vector within the Brillouin zone, so that $\mathbf{Q}=\mathbf{q}+\boldsymbol{\tau}$ where $\boldsymbol{\tau}$ is a reciprocal-lattice vector. The form factors can be expressed as follows:

$$F^{(s)}_{\mu\nu}(\mathbf{Q})=4\pi i^{l(\nu)-l(\mu)}\sum_{\eta}G^{\eta}_{\mu\nu}i^{-l(\eta)}Y_{\eta}(\hat{\mathbf{Q}})\int r^2dr\, j_{l(\eta)}(Qr)R_{l(\mu)}(r)R_{l(\nu)}(r) \ , \tag{6}$$

$$\mathbf{F}^{(o)}_{\mu\nu}(\mathbf{Q})=2\pi\left[\frac{4\pi}{3}\right]i^{l(\nu)-l(\mu)}\sum_{\xi}i^{-l(\xi)}Y_{\xi}(\mathbf{Q})\sum_{\eta}(A+B) \tag{7}$$

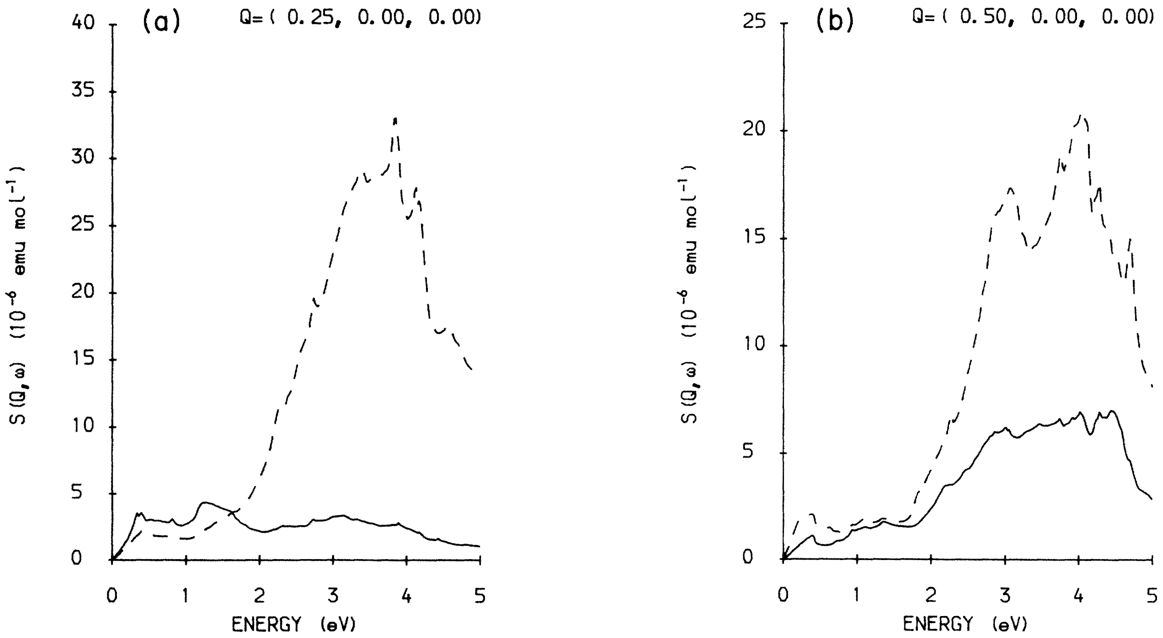

FIG. 1. Noninteracting $S(\mathbf{Q},\omega)$ for vanadium for values of $\mathbf{Q}$ in the [100] direction. Solid line is the spin part. Dashed line is the orbital part.

where

$$A = G^{\eta}_{\mu\nu} G^{\xi}_{(1m)\eta} \int \{ r^2 dr\, j_{l(\xi)}(Qr)(R_{l(\mu)} R'_{l(\nu)} - R_{l(\nu)} R'_{l(\mu)}) + \tfrac{1}{2}[l_\nu(l_\nu+1) - l_\mu(l_\mu+1)] r\, dr\, j_{l(\xi)}(Qr) R_{l(\mu)} R_{l(\nu)} \} , \tag{8}$$

$$B = \tfrac{1}{2}[G^{\eta}_{(1m)\mu} G^{\xi}_{\eta\nu} - G^{\eta}_{(1m)\nu} G^{\xi}_{\eta\mu}] l_\eta (l_\eta + 1) \int r\, dr\, j_{l(\xi)}(Qr) R_{l(\mu)} R_{l(\nu)} . \tag{9}$$

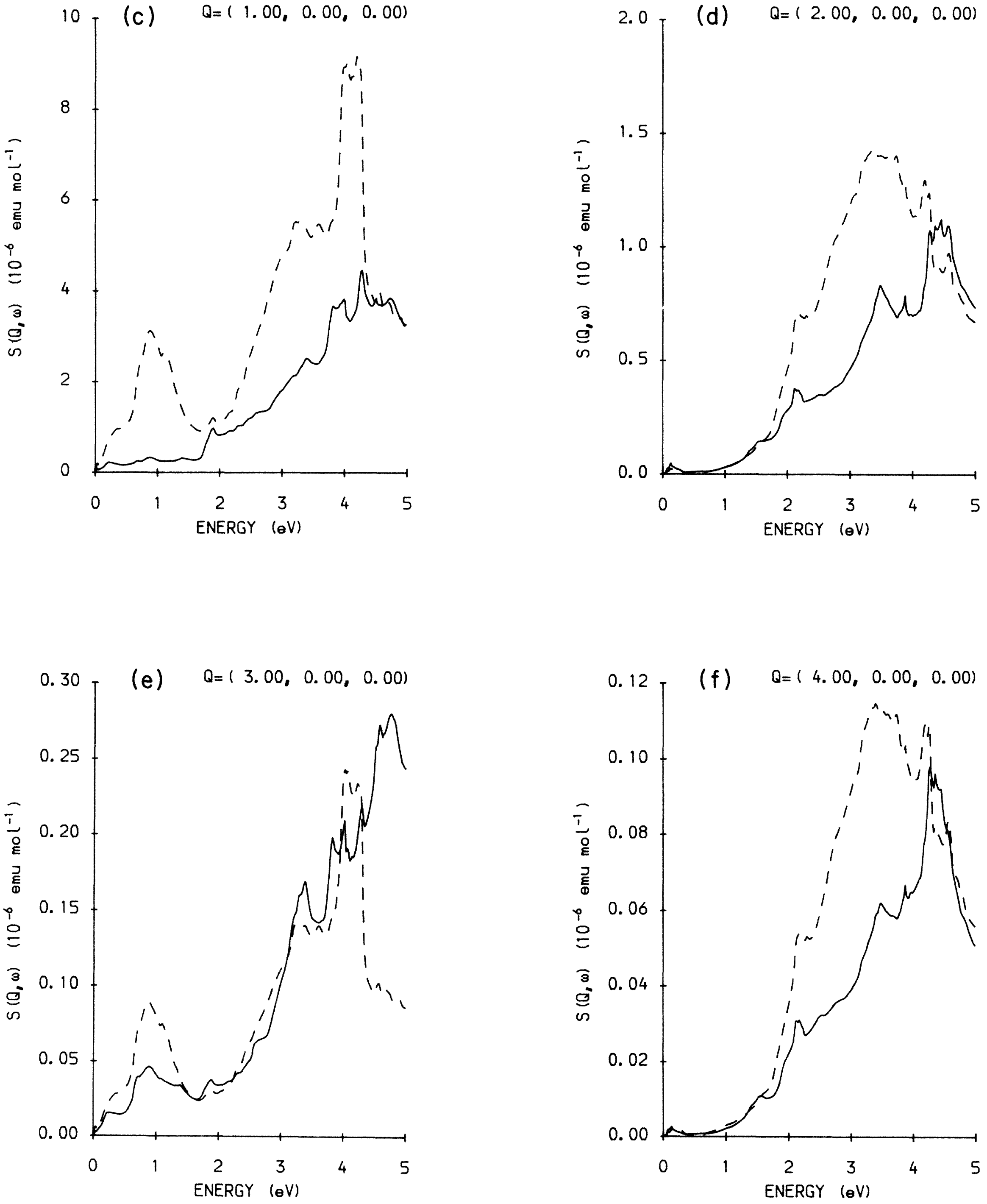

FIG. 1. (*Continued*).

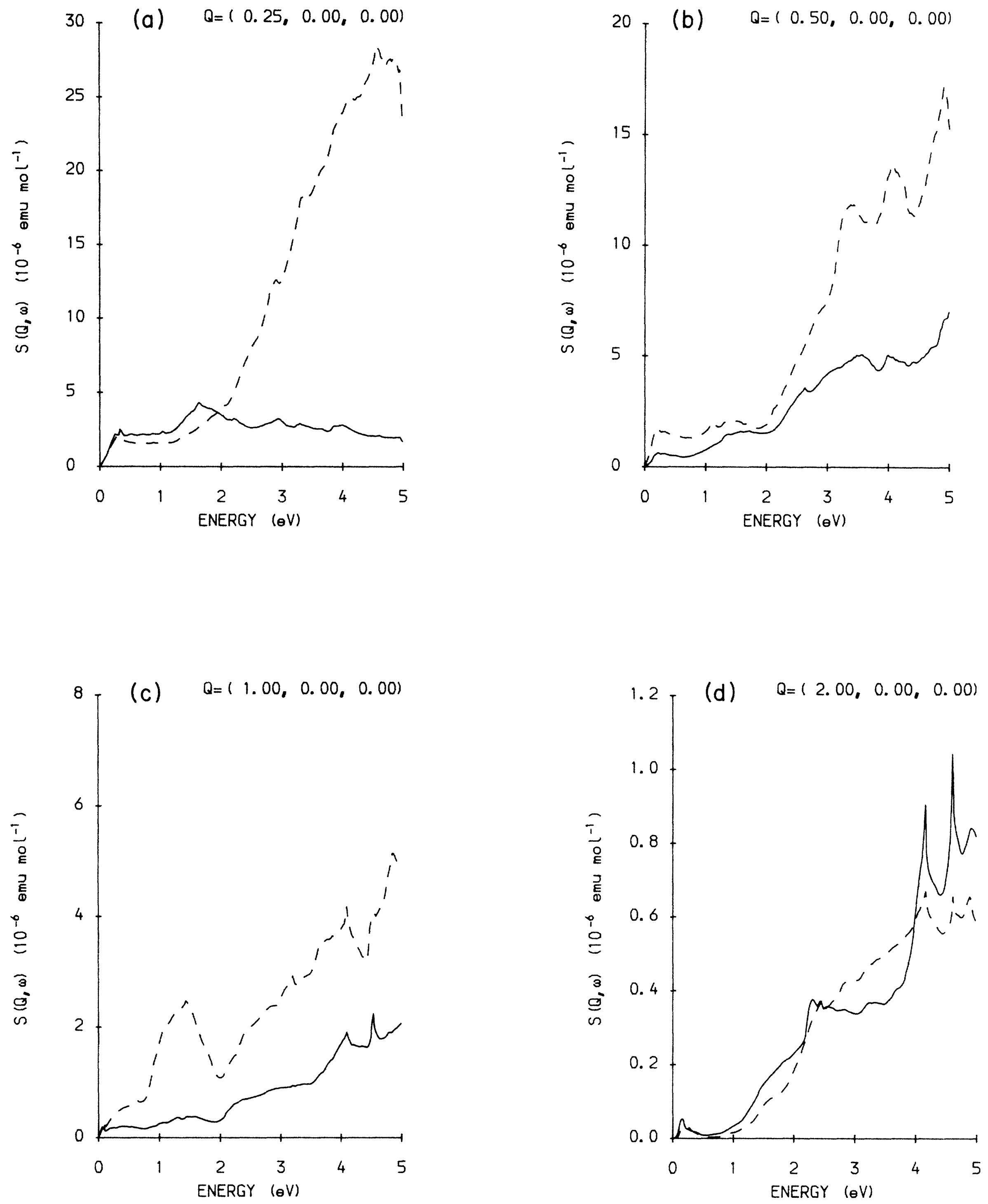

FIG. 2. As in Fig. 1, but for niobium.

In the above, $j_l(x)$ are Bessel functions of fractional order. $R_l' \equiv dR_l(r)/dr$ and the Gaunt coefficients are defined as

$$G^{\eta}_{\mu\nu} = \int d\hat{\mathbf{r}}\, Y_{\mu}(\hat{\mathbf{r}}) Y_{\nu}(\hat{\mathbf{r}}) y_{\eta}(\hat{\mathbf{r}}) \ . \tag{10}$$

The $x,y,z$ components of $\mathbf{F}^{(o)}_{\mu\nu}(\mathbf{Q})$ in (7) correspond to the values of $m$ of $1, -1, 0$ respectively, in $(lm)$.

The numerical calculations were performed for the cubic-symmetry paramagnets V, Nb, Rh, and Pd using a Korringa-Kohn-Rostoker (KKR) band-structure routine

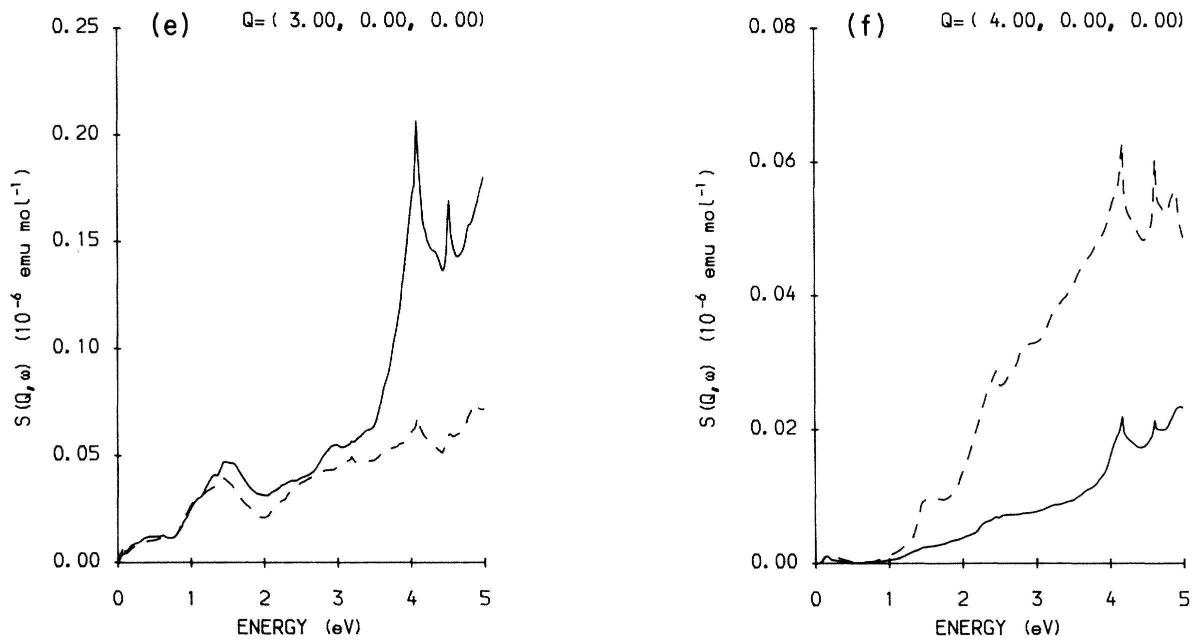

FIG. 2. (*Continued*).

and the self-consistently-derived potentials of Moruzzi *et al.*[10] The wave-function expansion (2) includes $s$-, $p$-, and $d$-symmetry terms. The zone sums in (3) were done by the tetrahedron method.[11] The radial integrals in (6), (8), and (9) were performed over the Wigner-Seitz sphere, the muffin-tin radial wave functions being extrapolated to the Wigner-Seitz radius.

## III. NUMERICAL RESULTS

The results of calculations of the unenhanced dynamical structure factor $S(\mathbf{Q},\omega)$ for bcc V and Nb and fcc Rh and Pd are shown in Figs. 1–4 for several values of $\mathbf{Q}$ in the [100] direction. $S(\mathbf{Q},\omega)$ is in units of $10^{-6}$

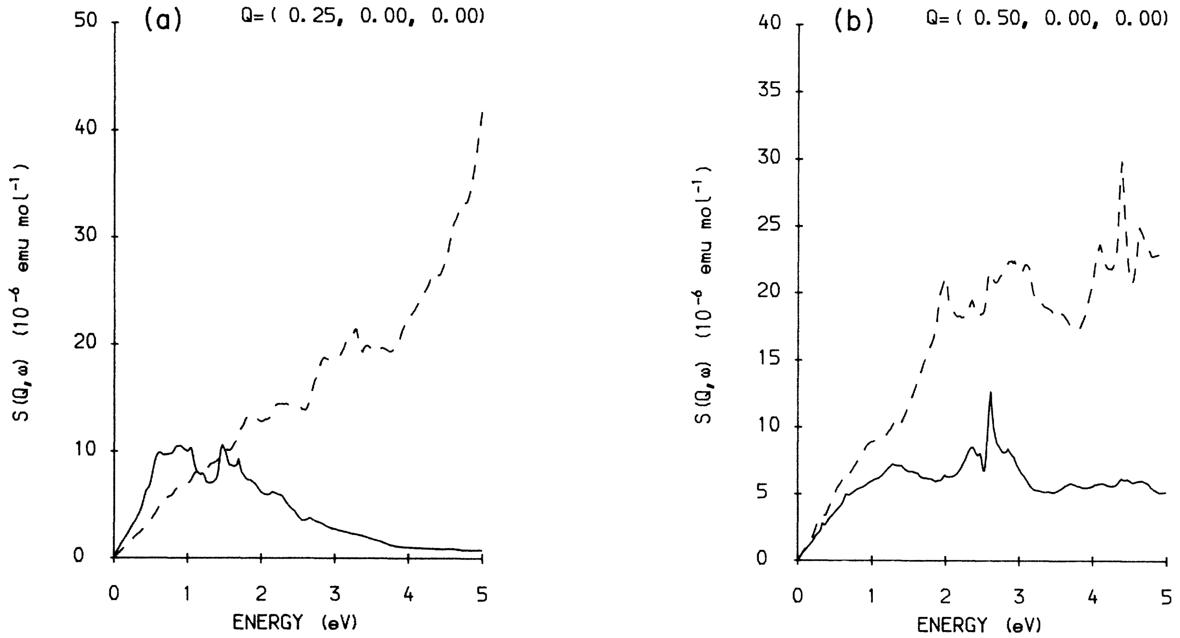

FIG. 3. As in Fig. 1, but for rhodium.

emu mol$^{-1}$ and the spin and orbital contributions are displayed separately by solid and dashed lines. $Q$ is in units of $2\pi/a$, which is 2.14, 1.92, 1.64, and 1.60 Å$^{-1}$ for V, Nb, Rh, and Pd, respectively.

The most striking feature of the results is the dominance of the orbital part of $S(\mathbf{Q},\omega)$ throughout a wide range of the parameter space $(\mathbf{Q},\omega)$. We also notice that there is no systematic decrease in the relative magnitude of the orbital part as compared to the spin part as higher values of $\mathbf{Q}$ are approached. In other words, the $Q^{-2}$ factor in (5) does not appear to dominate the behavior. This is particularly apparent in Pd (Fig. 4), where the comparative magnitude of the orbital and spin parts is roughly the same for both $Q=1$, and $Q=3$ (and also

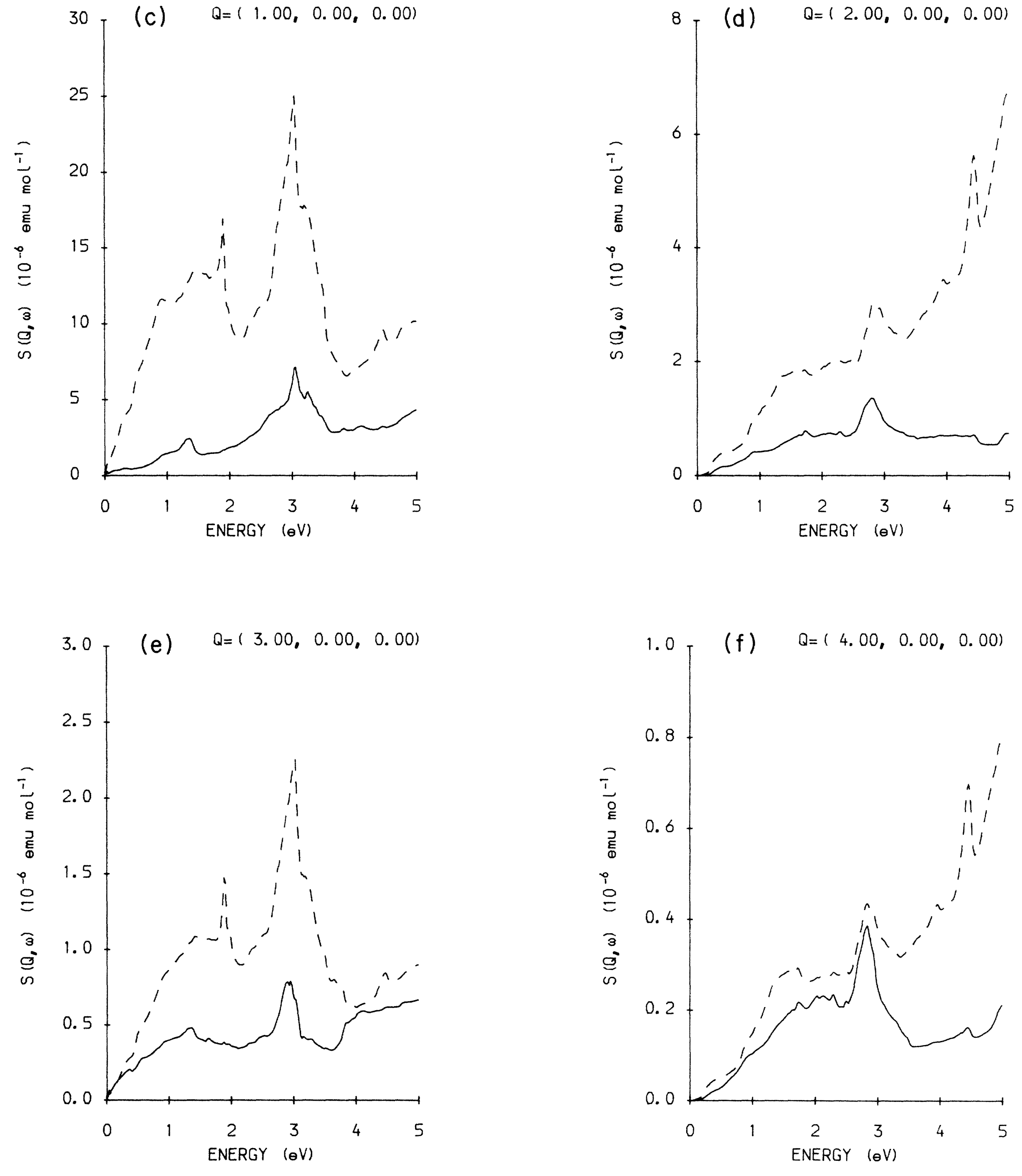

FIG. 3. (*Continued*).

$Q=2$ and 4). The trend is similar in Rh (Fig. 3), and in the $Q=2$ and 4 plots of V (Fig. 1), while in Nb (Fig. 2) the orbital contribution is relatively larger for $Q=4$ than it is for $Q=2$.

Features in the plots arising from the band structure tend to be more striking in the orbital part of $S(\mathbf{Q},\omega)$. The wave-vector transfer $\mathbf{Q}$ is related to a wave vector $\mathbf{q}$ in the Brillouin zone by the relation $\mathbf{Q}=\mathbf{q}+\boldsymbol{\tau}$. For both $Q=1$ and $Q=3$, for example, $\mathbf{q}$ is the same but the reciprocal-lattice vector is different. The details of the band structure enter through the Green's function $\Gamma$ in (4) and (5), and $\Gamma$ depends on $\mathbf{q}$. It is the form factor that depends on the full $\mathbf{Q}$. Referring to the $Q=1,3$ curves of Pd (Fig. 4), we can see the pronounced structural features in the orbital contribution while those in the spin part are much less prominent and more subject to modification

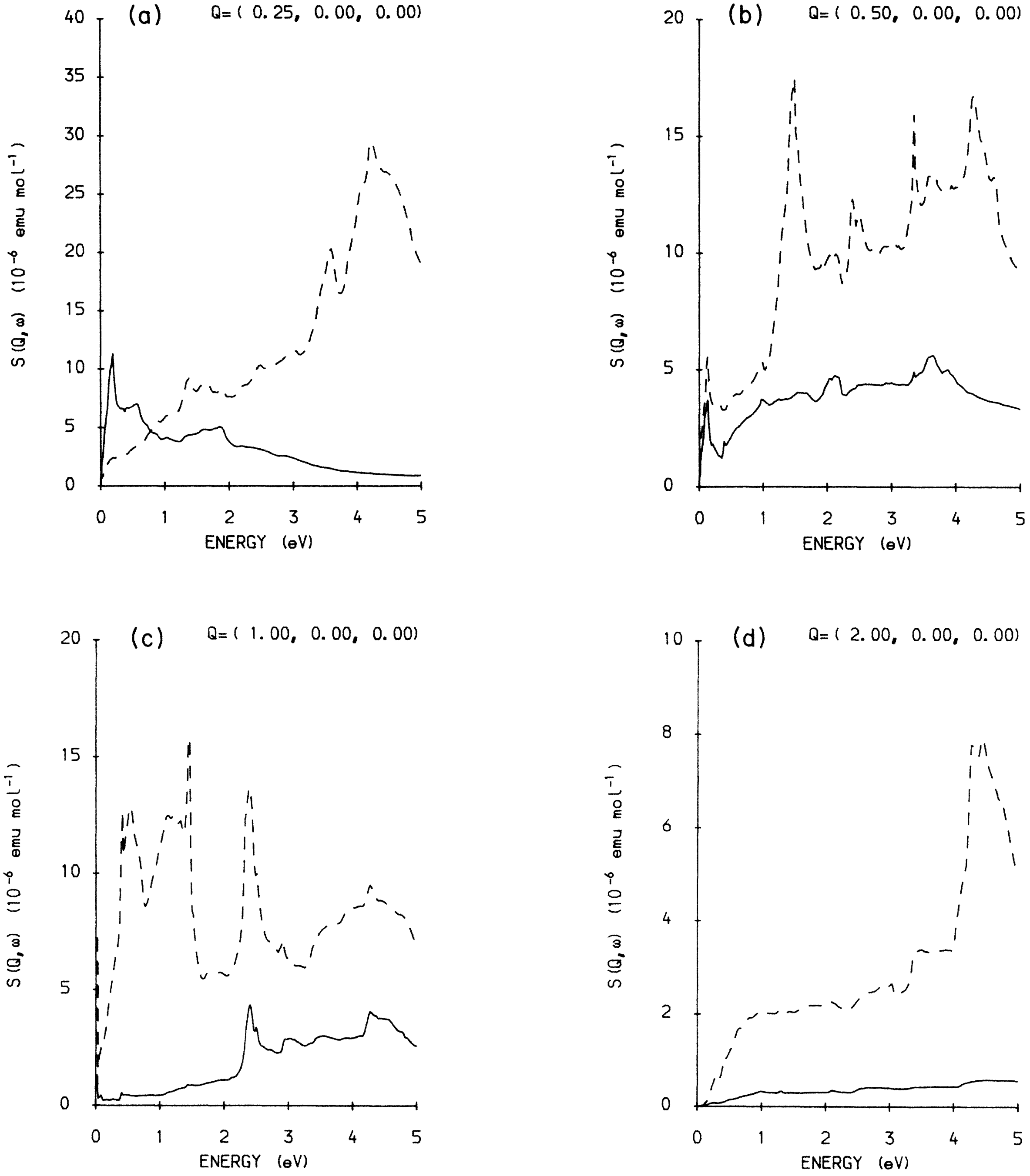

FIG. 4. As in Fig. 1, but for palladium.

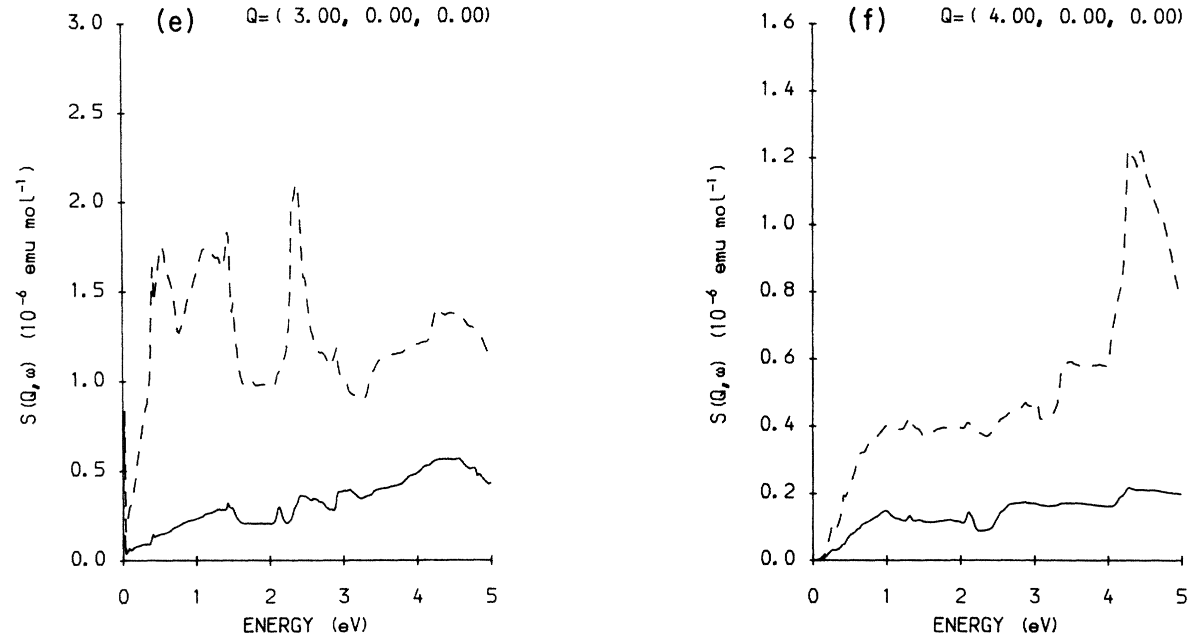

FIG. 4. (*Continued*).

from form-factor effects when going to a different reciprocal-lattice vector. For the bcc metals (Figs. 1 and 2) the feature in the orbital part at $1-1\frac{1}{2}$ eV for $Q=1$ and 3 is present also in the spin part at $Q=3$, but the form factor has eroded it at $Q=1$. It is also possible for structure to be present in the spin part but absent from the orbital part; Pd (Fig. 4) and Rh (Fig. 3) with $Q=0.25$ provide good examples of this.

Let us first understand why the $Q^{-2}$ factor in (5) appears to be ineffective in reducing the relative importance of the orbital contribution at high $Q$. Although the $s$ and $p$ contributions are included in the calculations of $S(\mathbf{Q},\omega)$, and $d$ components are dominant. For these $(l_\nu = l_\mu = 2)$, $A$ in (8) vanishes identically. For $d$ orbitals, the component in (9) with the lowest value of $l(\xi)$ is $l(\xi)=1$. This is likely to be the most important one. Us-

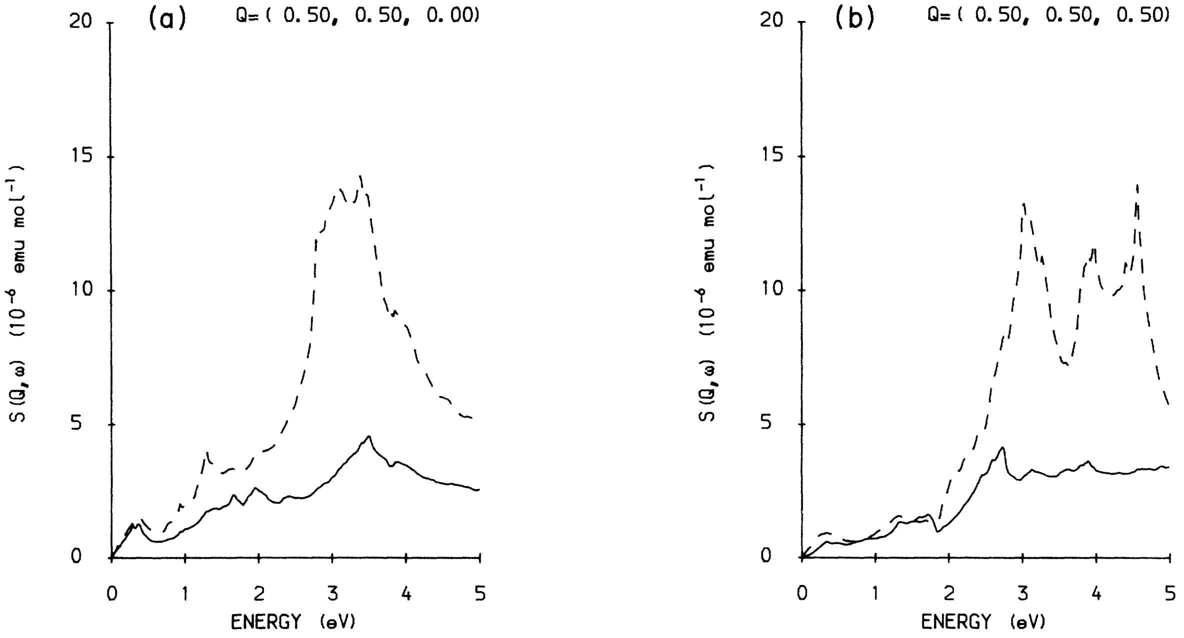

FIG. 5. Noninteracting $S(\mathbf{Q},\omega)$ for vanadium for values of $\mathbf{Q}$ associated with $(\frac{1}{2},\frac{1}{2},0)$ and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$. Solid line is the spin part. Dashed line is the orbital part.

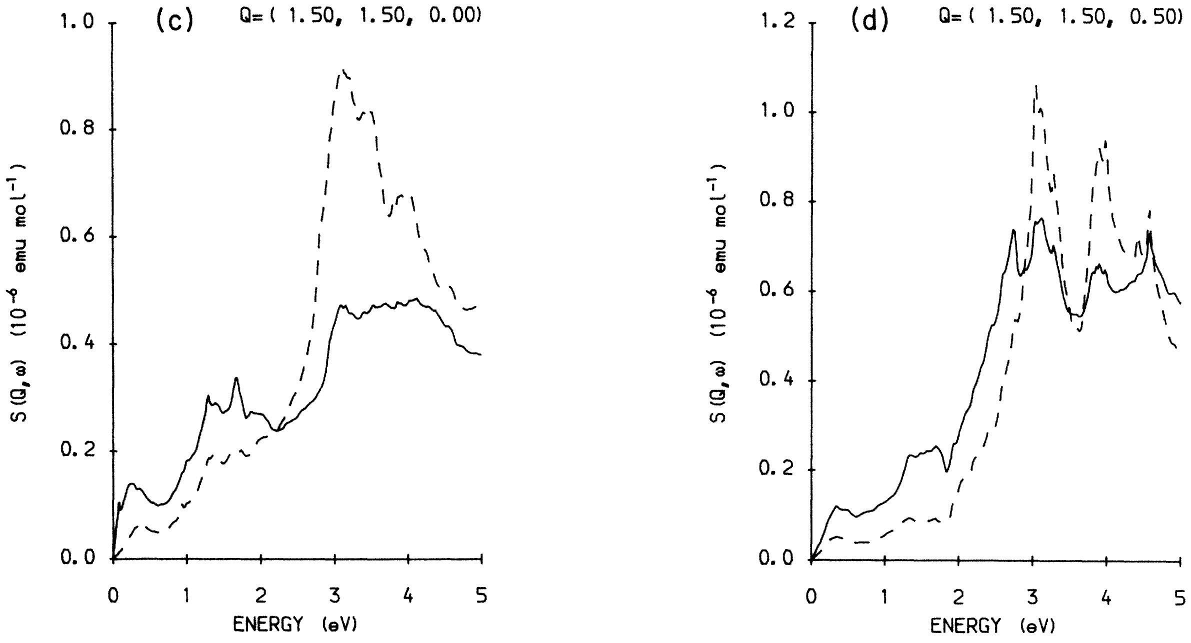

FIG. 5. (*Continued*).

ing a recurrence relation we can write

$$j_1(Qr)=\tfrac{1}{3}Qr[j_0(Qr)+j_2(Qr)]\ . \qquad (11)$$

Assuming that the dominant Bessel functions have low values of $l$, it can be seen that the most important radial integral in both the spin form factor (6) and the orbital one [(7) and (9)] is

$$\int r^2 dr\, j_0(Qr)[R_2(r)]^2\ .$$

In the orbital case, however, e.g., (11) introduces a factor $Q$ into the form factor which will give an exact cancellation with the $Q^{-2}$ in (5).

The second point to address is the dissimilarity that can occur between band-structure features appearing in

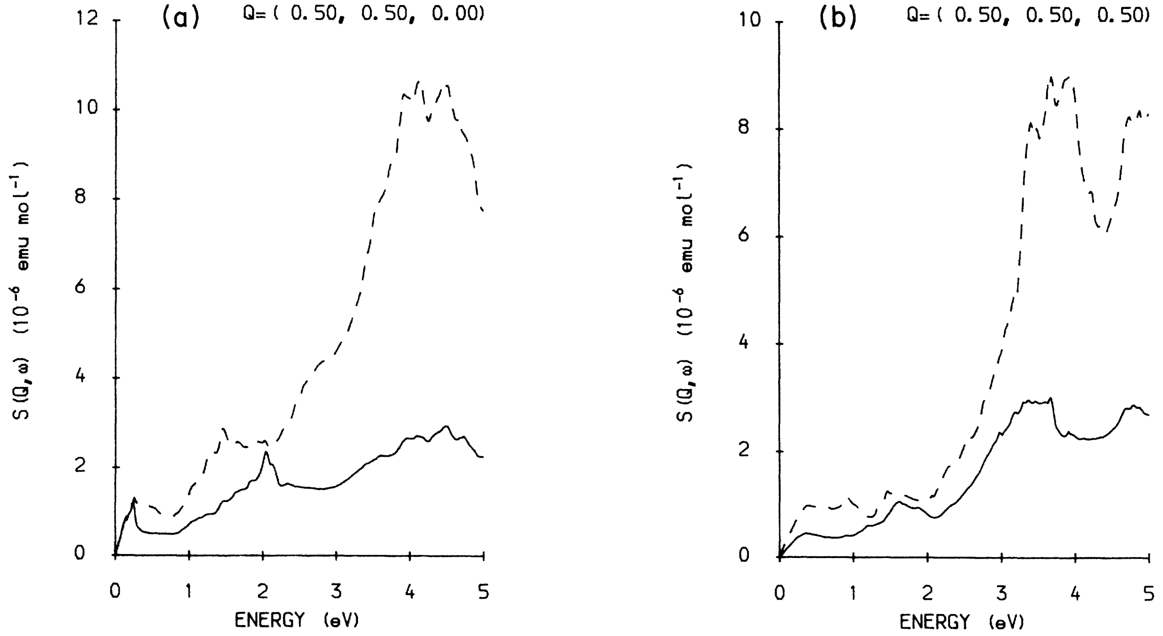

FIG. 6. As in Fig. 5, but for niobium.

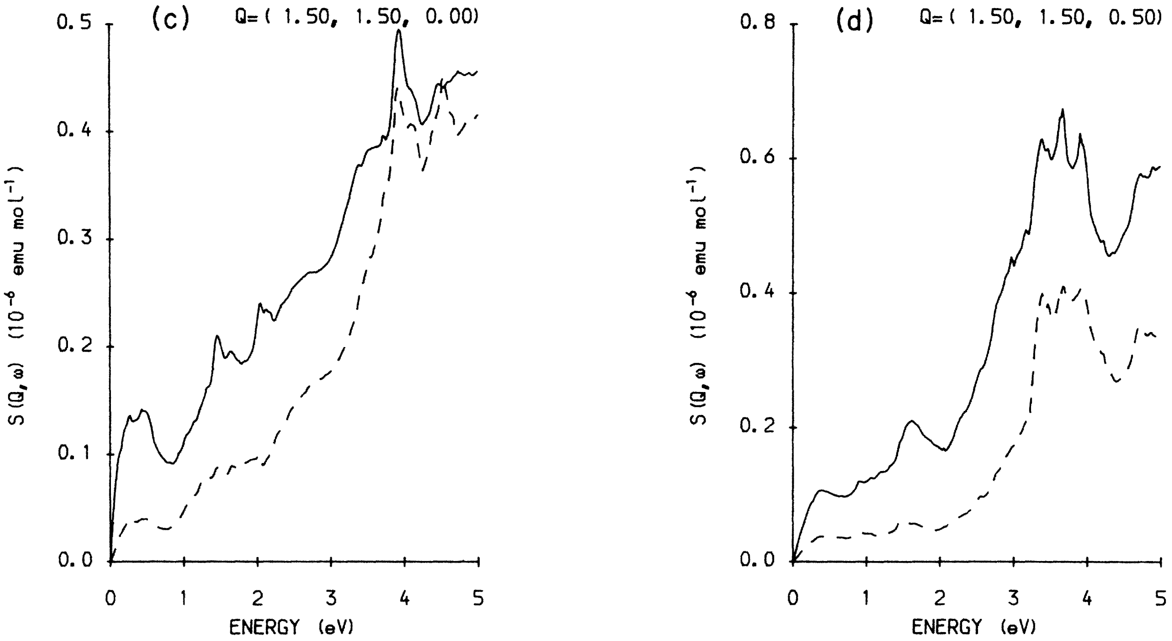

FIG. 6. (*Continued*).

the spin and orbital parts. For **Q** in the [100] direction, this can be understood in terms of simple selection rules applying to interband transitions. From a consideration of the Gaunt coefficients appearing in (6) and (7), it can be shown that the following inter-*d*-band transitions are *forbidden* for the [100] **Q** direction:

spin part, $e_g \rightarrow t_{2g}; t_{2g} \rightarrow e_g$;

orbital part, $e_g \rightarrow e_g$ .

Thus a feature arising from transitions between bands that both have a predominantly $e_g$ character will appear only in the spin contribution. Similarly, structure arising

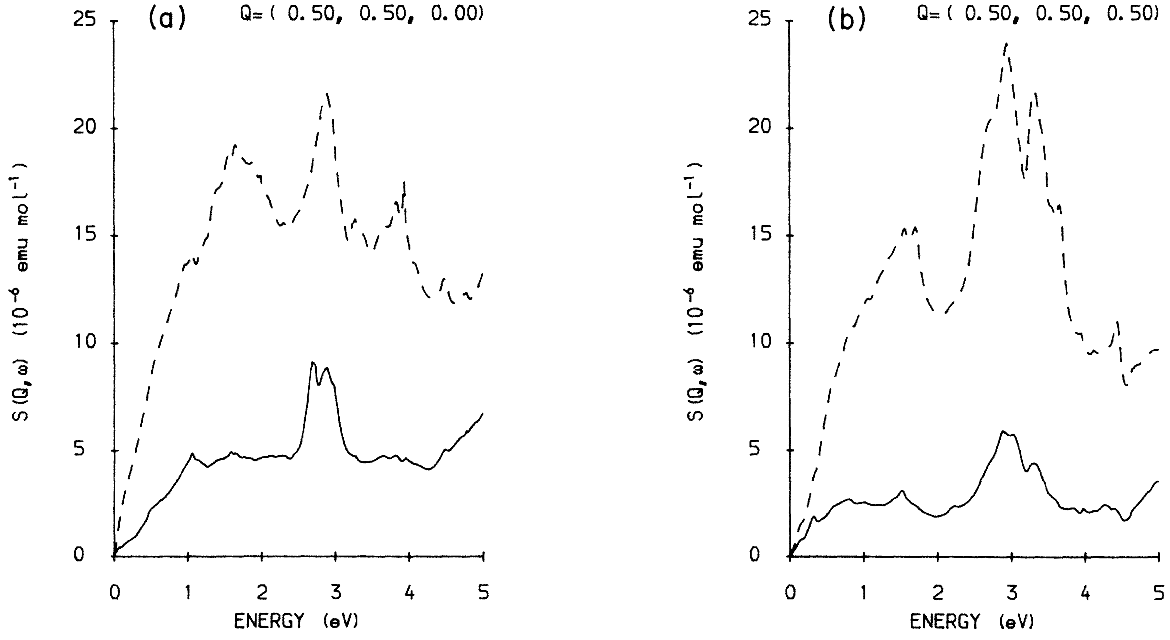

FIG. 7. As in Fig. 5, but for rhodium.

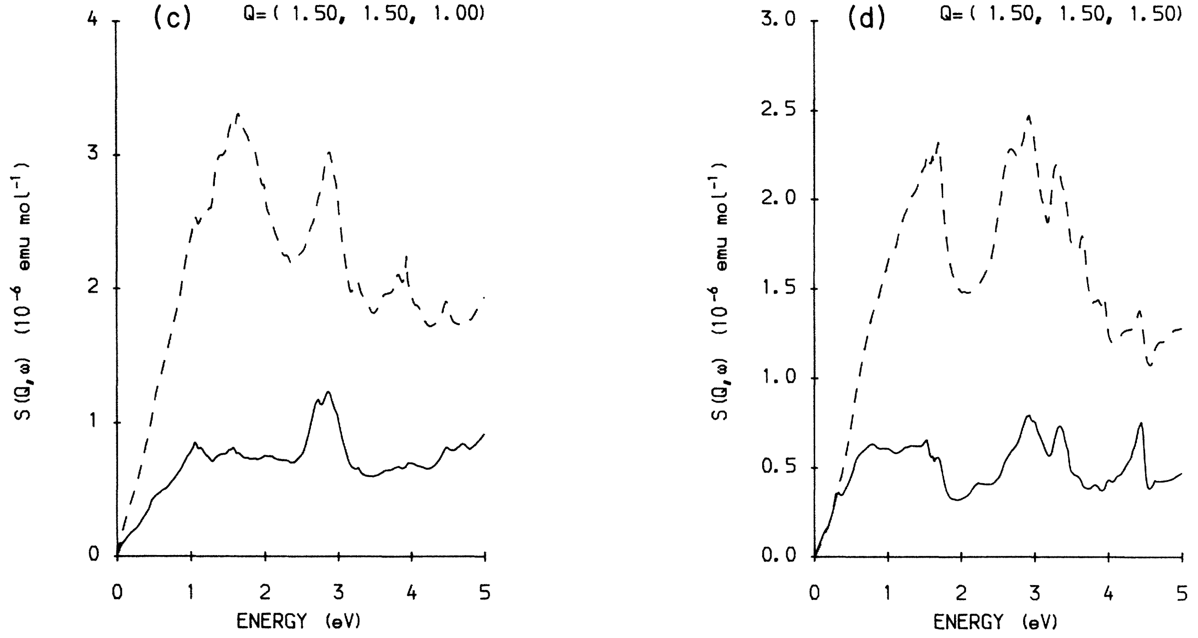

FIG. 7. (*Continued*).

from a transition from an $e_g$ band to a $t_{2g}$ band (or vice versa) will appear only in the orbital contribution. Furthermore, features that occur as a result of transitions between bands, each of which is of predominantly one symmetry type, will be rather insensitive to form-factor effects. The form factor will scale the plot without modifying its general shape. On the other hand, if the bands have a mixed symmetry, various cross terms occur and the form factor can weight these in differing ways for different reciprocal-lattice vectors, changing the appearance of the curve considerably.

Finally we show in Figs. 5–8 some results for the four metals with **Q** in other symmetry directions. Two values of **q** are considered: $(\frac{1}{2},\frac{1}{2},0)$ and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$; and two

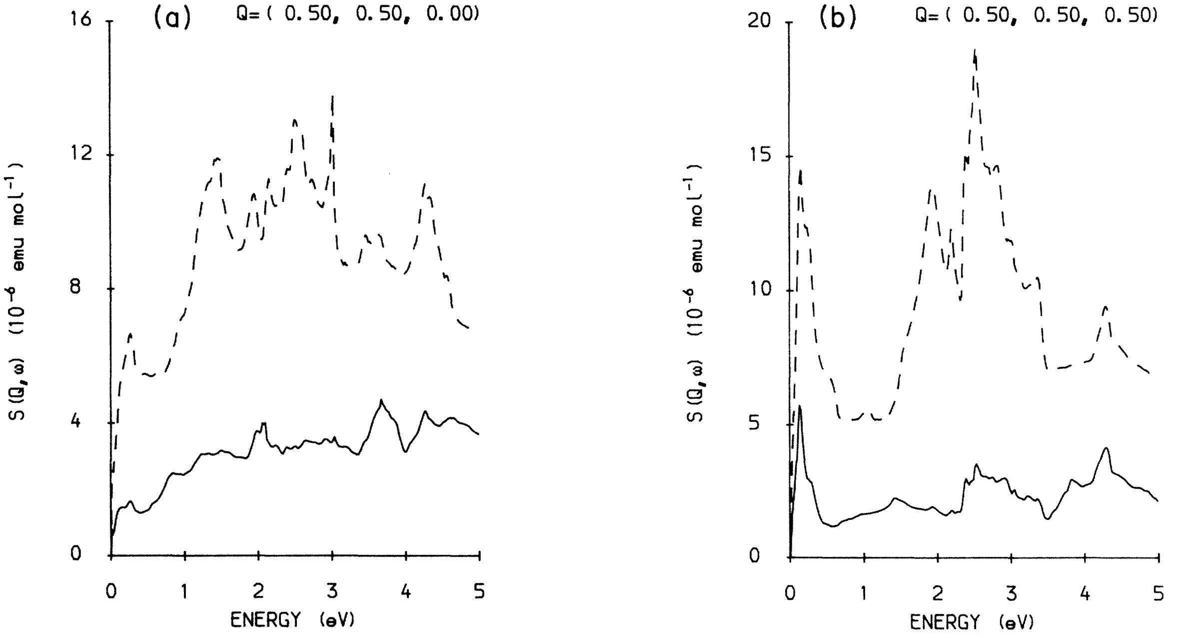

FIG. 8. As in Fig. 5, but for palladium.

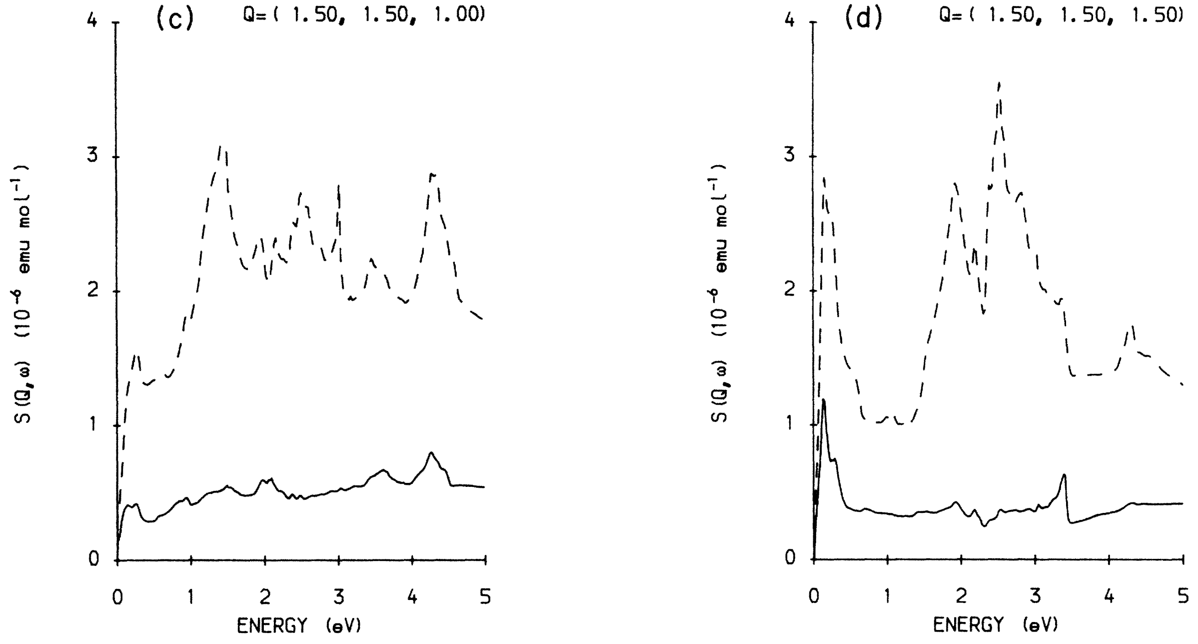

FIG. 8. (*Continued*).

reciprocal-lattice vectors $\tau$: (0,0,0) and, for bcc (1,1,0), and, for fcc (1,1,1). Similar observations to those made earlier apply here. There is no systematic decrease in the relative importance of the orbital part at higher Q and the structure in the orbital contribution is the more persistent when comparing plots at different reciprocal-lattice vectors. These points are particularly apparent in the fcc metals.

## IV. ENHANCEMENT EFFECTS

Stenzel and Winter[1] have calculated the enhancement to the spin susceptibility that arises from the exchange-correlation potential. They display numerical results for V and Pd for Q in the [100] direction. It is possible to discuss in broad terms the consequences of enhancement on our results for V (Fig. 1) from their findings.

Enhancement is not important for small $q$ (below $\sim 0.3$) and large $\omega$ (above $\sim 0.3$ eV), or for any value of $q$ if $\omega$ is larger than $\sim 3$ eV. However, for $q$ near the zone boundary, enhancement can increase the spin susceptibility by an order of magnitude at energy transfers of $\sim 1.5$ eV, although factors of 3–5 are more widespread.

Now consider the effect of this behavior on the plots in Fig. 1. The predominance of the orbital contribution in the $Q = 0.25$, 2, and 4 plots will remain unchanged except at the lowest energy transfers (below $\sim 0.1$ eV). For the other three plots, the relative importance of the spin and orbital contributions above $\sim 3$ eV will remain unchanged, while the low-energy spin part will be enhanced by a factor of about 4 (or $\sim 10$ around 1.5 eV for the $Q = 1$ and 3 plots). Analogous behavior will occur in the other materials although obviously the details will depend on the metal. Enhancement effects when they do occur will be particularly pronounced in Pd, of course.

The question of enhancement to the orbital susceptibility remains to be addressed. The orbital contribution is related to a density-density correlation function and hence has features in common with the inverse dielectric function. For example, we expect that any enhancement effects will arise from the direct rather than the exchange interaction and they will be associated with a plasmon pole. As a consequence of the orbital form factor, however, this pole can be shown to have zero weight, at least in the $\mathbf{q} \rightarrow 0$ limit. Because of this we expect that many-body corrections will be less important for the orbital part than for the spin part.

## V. CONCLUSIONS

The purpose of this paper has been to assess the relative importance of the orbital contribution to $S(\mathbf{Q},\omega)$ for the $d$-band transition metals. As far as the unenhanced susceptibility is concerned, the orbital part clearly gives a major contribution over a wide range of energy and wave-vector transfers. We have made tentative predictions about the way in which both the spin and orbital parts will be affected by the electron-electron interaction. This is potentially a subtle issue, however, and detailed calculations would be of considerable interest. We plan to report on this matter at a later stage.

## ACKNOWLEDGMENTS

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