Inelastic scattering of x rays at intermediate momentum transfers

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The dynamical structure factor of the K-shell electron of Cu at intermediate momentum transfers is calculated on the basis of both the one-electron and the one-site approximation. The aim is to study the structure of the profile found experimentally by Namikawa and Hosoya [Phys. Rev. Lett. **53**, 1606 (1984)] under the condition $qa \approx 1$, where a is the orbital radius of the K-shell electron of Cu and q is the photon momentum transfer. The structures observed by Namikawa and Hosoya experimentally or those obtained theoretically on the basis of the single-orthogonalized-plane-wave approximation are not found in the present calculation. Indeed, the present theoretical result agrees with the spectral shape obtained experimentally by Manninen, Hämäläinen, and Graeffe [Phys. Rev. B **41**, 1224 (1990)] and revises the calculation based on the impulse approximation.

I. INTRODUCTION

The spectral shape for the scattering of γ rays (59.57 eV) by K-shell electrons of Cu has been obtained by Namikawa and Hosoya[1] (NH) with use of a coincidence method. The purpose of the experiment was to determine the spectral shape in the intermediate-momentum-transfer region $qa \approx 1$, where q is the photon momentum transfer and a is the radius of the inner-shell electron under consideration. For a scattering angle $\theta = 135°$, the observation of two peaks named peak R and peak DT near the threshold was claimed, as was another peak named peak C near 40 keV.

On the other hand, Manninen *et al.*,[2,3] after performing an experiment similar to that done in NH, reported preliminary spectra different from those of NH, although two peaks, near the threshold and near 40 keV, have been found. Their results, as they themselves admit, still contained contributions from false coincidences so that further investigation on the profile would be required.

Using syncrotron radiation, Marchetti and Franck[4,5] obtained a spectral shape for the scattering of γ rays with incident energies 70 and 62 keV and concluded that there were no structures near threshold. They reported a spectral shape which agrees with the theoretical calculation obtained by atomic wave functions for both the initial and final states. The spectra obtained, however, did not have sufficient resolution to provide a conclusive answer to this problem.

Very recently, Manninen *et al.*,[6] based on another experiment on the scattering of γ rays (59.537 keV) by K-shell electrons of Cu and Zr, concluded that there is no two-peak structure near threshold. They reported that the spectra obtained agree very well with the corresponding theoretical result based on the impulse approximation.

Since the impulse approximation is valid for scattering under the condition $qa \gg 1$,[7,8] the agreement between the theoretical result based on the impulse approximation and the experimental result done under the condition $qa \approx 1$ deserves further scrutiny. As already noted, an analysis based on an atomic model of the wave function in the same energy region also claimed to be in agreement with the corresponding experimental result.[5] We have another example of scattering by K-shell electrons of Li under the condition $qa \approx 1$, where the spectral shape changes considerably from that obtained by the impulse approximation.[9-11] The purpose of this paper is to report our recent theoretical result on this problem.

II. FORMULATION

The spectral shape for the present problem in the one-electron approximation (in units in which $\hbar = 1$, $e^2 = 2$, and $m = \frac{1}{2}$), is given by a standard formula[7-10]

$$w(\omega,\theta,\phi)d\omega\, d\Omega = \frac{8n_0}{c^3}\frac{N}{V}(1+\cos^2\theta) \times \frac{\omega_1}{\omega_0} S(\mathbf{q},\omega)d\omega\, d\Omega \ , \tag{1}$$

where

$$S(\mathbf{q},\omega) = 2 \sum_f^{(\text{unoccupied})} \left| \int \psi_f^* \exp(-i\mathbf{q}\cdot\mathbf{r})\psi_{1s} d\mathbf{r} \right|^2 \times \delta(\omega - \varepsilon_f + \varepsilon_{1s}) \ , \tag{2}$$

and

$$\mathbf{q}=\mathbf{k}_0-\mathbf{k}_1, \quad \omega=\omega_0-\omega_1 . \tag{3}$$

Here, $\mathbf{k}_0$ and ω_0 are, respectively, the wave-number vector and the energy of the incident photon; $\mathbf{k}_1$ and ω_1 are those of the outcoming photon. ψ_{1s} and ε_{1s} are the wave function and the energy of the electron in the $1s$ state, while ψ_f and ε_f are those in the final state. The summation is taken over the unoccupied states f.[8,9]

In our previous paper[12–15] we started analysis based on a single orthogonalized plane wave (OPW) for the wave function in the final state, where we could take into account the orthogonality to the core parts and the periodicity in the wave function. Although the spectral shape is quite different from that obtained by NH, we found two peaks which could be identified as peak R and peak C, following NH. These two peaks have been interpreted as a manifestation of the complicated behavior of the wave function near the K shell which should be revealed for the scattering under the condition $qa \approx 1$; with peak R being due mainly to the $1s$ part and peak C, mainly to the plane-wave part.[12,13]

In our present calculation to reexamine the spectral shape, we use the one-site approximation,[9,10] taking into account the variation of the potential $V(r)$ in the final state due to the presence of the inner-shell hole. The potential that we adopted is

$$V(r)=\begin{cases} -\dfrac{2Z}{r}+V_C(r)+v_{\rm ex}(r)-V_{\rm av}, & r<r_m \\ 0, & r>r_m , \end{cases} \tag{4}$$

where r_m is half the distance between the centers of the nearest-neighboring atoms of Cu. In Eq. (4), the first term is the Coulomb part of the nucleus with Z equal to 29. $V_C(r)$ is the direct Coulomb part of the inner-shell electrons that is obtained by solving Poisson's equation by use of the double-ζ functions of Clementi and Roetti.[16] $V_{\rm ex}(r)$ is the exchange Coulomb term estimated by the $X\alpha$ method of Slater for which the correlation-exchange parameter α is chosen to be 0.7. $V_{\rm av}$ is the average of the first three terms on the right-hand side of Eq. (4) in the region of $r_m<r<r_{\rm WS}$, representing the mean field potential due to the conduction electrons, where $r_{\rm WS}$ is the radius of the Winger-Seitz atomic sphere.

Since $V(r)$ [Eq. (4)] is spherically symmetric we may solve the Schrödinger equation by assuming a product wave function

$$\psi_{klm}(\mathbf{r})=C_{kl}R_{kl}(r)Y_{lm}(\theta,\phi) , \tag{5}$$

with energy given by

$$\varepsilon_k=k^2=\omega-\varepsilon_{1s} , \tag{6}$$

where C_{kl} is the normalization constant to be given below and k is the wave number of the electron in the final state. In Eq. (6) the principle of energy conservation has been invoked.

The equation for $rR_{kl}(r)$ is given by

$$\left[-\frac{d^2}{dr^2}+V(r)+\frac{l(l+1)}{r^2}\right][rR_{kl}(r)]=\varepsilon_{kl}rR_{kl}(r) . \tag{7}$$

Equation (7) is solved numerically in the region of $r<r_m$ and the solution is continued to the region of $r>r_m$ in the form of

$$rR_{kl}(r)=kr[j_l(kr)\cos\delta_{kl}-n_l(kr)\sin\delta_{kl}] , \quad \text{for } r>r_m , \tag{8}$$

where $j_l(kr)$ and $n_l(kr)$ are the spherical Bessel functions. We normalize the wave function in a very large sphere of radius R, obtaining C_{kl} in Eq. (5) independently of k and l as

$$C_{kl}=\left[\frac{2}{R}\right]^{1/2} . \tag{9}$$

In the final state, the summation over the quantum number m in Eq. (5) gives a factor $(2l+1)$ and the summation over k is replaced by

$$\sum_k \rightarrow \frac{R}{\pi}\int dk . \tag{10}$$

Then the spectral shape is given by a summation over the angular momentum l as

$$S(\mathbf{q},\omega)=\sum_l S^l(\mathbf{q},\omega) , \tag{11}$$

$$S^l(\mathbf{q},\omega)=\frac{2}{\pi k}(2l+1)\left[\int rR_{kl}(r)j_l(qr)rR_{1s}(r)dr\right]^2 , \tag{12}$$

where $R_{1s}(r)$ is the radial part of the $1s$ orbital.

III. RESULTS OF CALCULATION

The spectral shapes obtained with the one-site approximation from each partial wave $l=0,1,2$, and 3 are shown in Fig. 1. We have neglected the contributions from

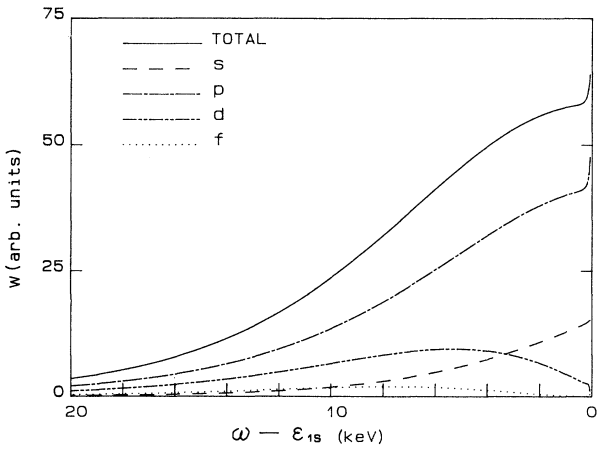

FIG. 1. The spectral shape obtained by the one-site approximation. Contributions from s, p, d, and f waves are shown, together with the total.

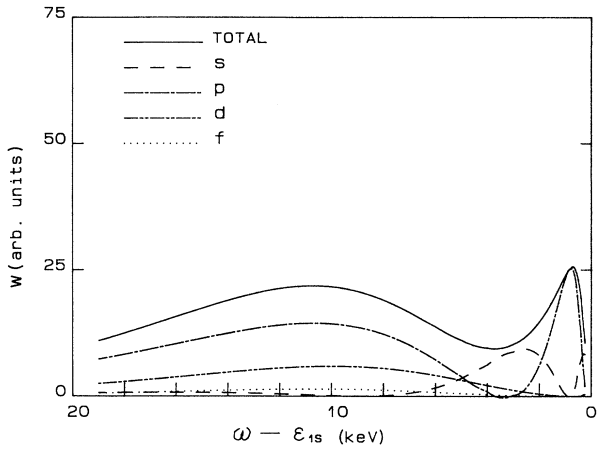

FIG. 2. The spectral shape obtained by a single OPW. Contributions from s, p, d, and f waves are shown, together with the total.

those with $l \geq 4$. As expected, the contribution from the $l=1$ partial wave is the largest in the whole spectra. Profiles for the $l=0$ and $l=1$ partial waves decrease monotonically from threshold while that by the $l=2$ partial wave has a maximum at 6 keV away from threshold. The total profile, given by the solid line in Fig. 1, decreases monotonically from threshold since the $l=2$ partial-wave contribution is not large.

Spectral shapes under the OPW approximation for each of l are shown in Fig. 2. A solid line gives the total profile which has a dip near 3 keV away from threshold. This is due to the decrease of the contribution of the $l=1$ partial wave in this region, which, however, has not been observed in recent experimental results.[5,6] The difference between the present result and the previous OPW result[13,15] is due to the difference in behavior of the wave functions in the region $r<0.2$, in which the K-shell electron is localized.

Figures 3 and 4 show clearly the difference between these functions; given are wave functions of electrons in the final state corresponding to spectra away from threshold 0.2 keV [Figs. 3 and 4(a)], 0.8 keV [Fig. 4(b)], and 3 keV [Fig. 4(c)]. In Fig. 3 we observe that the amplitude

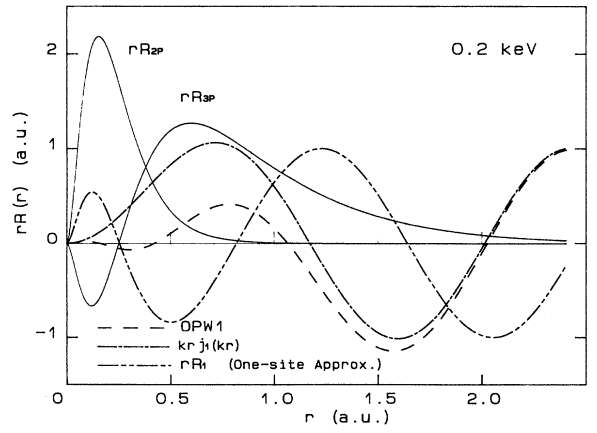

FIG. 3. Radial parts of wave functions in the single OPW, plane wave, one-site, and the impulse approximations, giving the portion in the spectra 0.2 keV away from threshold.

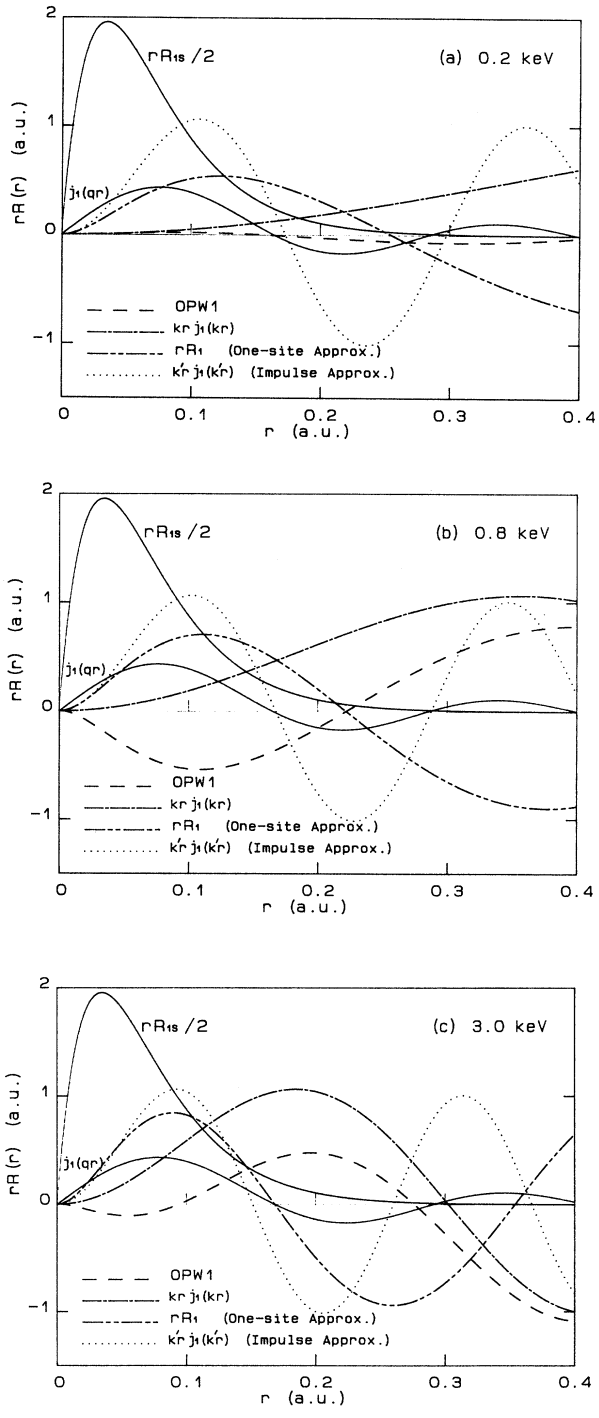

FIG. 4. Radial parts of wave functions in the single OPW, plane wave, one-site, and the impulse approximations, giving the portion in the spectra 0.2 keV (a), 0.8 keV (b), and 3 keV (c) away from threshold, respectively.

TABLE I. Values of parameters available to check the orthogonality of wave functions to the core parts for various electron energies η_{np} for the one-site approximation and ξ_{np} for a plane-wave approximation. See the text for the definitions.

Electron energy	200 eV	500 eV	800 eV	3 keV
η_{2p}	$+2.1\times10^{-2}$	$+2.1\times10^{-2}$	$+1.7\times10^{-2}$	$+4.4\times10^{-3}$
η_{3p}	-9.8×10^{-2}	-2.7×10^{-2}	-1.4×10^{-2}	-4.1×10^{-3}
ξ_{2p}	$+2.0\times10^{-1}$	$+3.3\times10^{-1}$	$+3.7\times10^{-1}$	$+2.6\times10^{-1}$
ξ_{3p}	$+5.5\times10^{-1}$	$+1.3\times10^{-1}$	-2.7×10^{-2}	-1.1×10^{-1}

of the OPW function is very small in the region $r<0.2$, giving a small contribution to spectra of the corresponding part in $S^l(\mathbf{q},\omega)$. The overlap of the function $j_1(kr)$ and the inner-shell orbital $R_{3p}(r)$ is large in this energy region, leading to a large deviation of the OPW function from $j_1(kr)$. On the other hand, the function $R_{k1}(r)$ of the present one-site approximation is pulled in considerably to have the corresponding first peak of $j_1(kr)$ in the region $r<0.2$. Figure 4(a) shows the function of the same energy as in Fig. 3 in the region $r<0.4$.

In Fig. 4(b) the OPW result and that obtained with the one-site approximation are similar; in Fig. 4(c), they are different because the requirement of orthogonality to the $2p$ has a dramatic effect on the OPW, making it depart from the wave function obtained under the one-site approximation.

To check the orthogonality of the function $R_{k1}(r)$ to the inner-shell orbitals $R_{np}(r)$ we calculated η_{np} and ξ_{np} defined as

$$\eta_{np}=\int dr\, r^2 R_{k1}(r)R_{np}(r)\ , \tag{13}$$

$$\xi_{np}=\int dr\, r^2 k j_1(kr)R_{np}(r)\ . \tag{14}$$

Here we have used the wave functions obtained by Clementi and Roetti[16] for inner-shell orbitals. Table I shows that all η_{np}'s are small and the required orthogonality is well satisfied.

IV. DISCUSSION

We may conclude that, for the present problem with $qa\approx1$, the effect of the potential on the wave function is dominant in pulling in the free-electronlike wave function rather than in deforming it appreciably. This may be the reason why both calculations based on the atomic and the impulse approximation give similar results. In the impulse approximation, the wave number k' is chosen so that $k'rj_1(k'r)$ is well fitted to $rR_{k1}(r)$ of the one-site approximation.

In applying the present result to the problem of solids we notice that the wave function of the one-site approximation does not satisfy Bloch's periodicity condition contrary to the case of the OPW, but it took account of the effect of the local field sufficiently. The wave function could work as a basis function to start with for such a band calculation as the augmented-plane-wave method[17] or the Green's-function method.[18] By imposition of periodicity, the characters of the wave functions are mixed and the relative contribution of the $s,p,d,\ldots$, parts of the wave function changes with the energy of the scattered electrons, resulting in a change in the spectral shape, but still not like that found by NH or theoretically by the single-OPW approximation. This will be discussed in a forthcoming paper. Our conclusion is that the structure near threshold found by NH may be caused by other processes irrelevant to $S(\mathbf{q},\omega)$, e.g., a sequential scattering process as suggested by Manninen,[2,3] i.e., first elastic scattering in the sample and then Compton scattering in the x-ray detector.

According to the recent experimental result of Manninen *et al.*[6] the spectra decrease monotonically from threshold. The theoretical result based on the impulse approximation agrees well with their experimental result. The spectrum obtained under the one-site approximation has a weaker energy dependence, agreeing better with the experimental result[6] as shown in Fig. 5. We see a clear difference between the result based on the impulse approximation and that based on the one-site approximation. Our result agrees fairly well with the result based

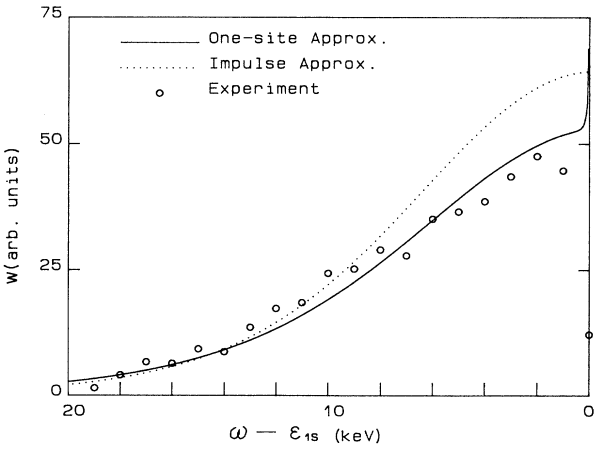

FIG. 5. The spectral shape by the one-site approximation (solid line) compared with the experimental result (circles) of Manninen *et al.* [Phys. Rev. B **41**, 1224 (1990)].

on an atomic wave function of Marchetti and Franck.[5] If we take account of the periodicity of the crystal in the wave function, the whole spectral shape is made flatter than the one presented here, thus agreeing better with the experimental result of Manninen *et al.*[6] Details of this work will be published elsewhere.

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