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Determination of the angular distribution of the local density of free-electron states in low-*Z*-atom crystals using inelastic x-ray scattering spectra

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Abstract. For the first time, an experiment for determining the angular distribution of the local density of free-electron states in low-*Z*-atom crystals is suggested. This experiment is based on inelastic x-ray scattering (IXS) spectroscopy associated with low-*Z*-atom *K*-level excitations of solids. On the basis of the method developed for the calculation of the double-differential cross section of the IXS, a simple expression for nondiagonal matrix elements of the Green function is obtained.

Information on the local density $\rho(\mathbf{r}, \varepsilon)$ of unoccupied electron states (LDOS) in solids is of great importance in studies of chemical bonding and local atomic structure in crystals and amorphous media. The imaginary part of the one-electron Green function $G^+(\mathbf{r}, \mathbf{r}', \varepsilon)$ (GF) [1] contains all the information about the LDOS:

$$\rho(\mathbf{r}, \varepsilon) = \sum_n |\varphi_n(\mathbf{r})|^2 \delta(\varepsilon - \varepsilon_n) = -\frac{1}{\pi} \text{Im} G^+(\mathbf{r}, \mathbf{r}, \varepsilon). \quad (1)$$

In the muffin-tin approach for GF calculation [2, 3] the expression for the LDOS $\rho(\mathbf{r}, \varepsilon)$ near a given atom has the form

$$\rho(\mathbf{r}, \varepsilon) = \sum_L \sum_{L'} F_{LL'}(\varepsilon) R_L(\mathbf{r}, \varepsilon) R_{L'}(\mathbf{r}, \varepsilon) Y_L(\Omega) Y_{L'}(\Omega) \quad (2)$$

and for the spherically averaged LDOS $\rho(r, \varepsilon)$ has the form

$$\rho(r, \varepsilon) = \frac{1}{4\pi} \oint \rho(\mathbf{r}, \varepsilon) d\Omega = \frac{1}{4\pi} \sum_L F_{LL}(\varepsilon) R_L^2(r, \varepsilon) \quad (3)$$

where $R_L(r, \varepsilon)$ is the regular solution of the radial Schrödinger equation, $Y_L(\Omega)$ is a real angular harmonic, and $F_{LL'}$ depends on the normalization of $R_L(r, \varepsilon)$. If we normalize $R_L(r, \varepsilon)$ as in [4, 5], then F_{LL} is to be interpreted as the number of electron states with spherical symmetry L per unit energy interval.

One can see from (3) that the diagonal elements F_{LL} give us complete information on the spherically averaged LDOS $\rho(r, \varepsilon)$ near the atom investigated, but the angular distribution of the LDOS $\rho(\mathbf{r}, \varepsilon)$ (equation (2)) is determined by both diagonal and nondiagonal elements of the matrix of elements $F_{LL'}$ ($L \neq L'$).

Various experiments (such as XAS studies [2, 3]) have been performed which have produced information on some matrix elements of GF, but unfortunately these experiments do not provide in principle complete information on the GF. In fact, XAS with the photon

energy near the K absorption edge provides information on the matrix elements $F_{lm,lm'}$, while XAS near the $L_{2,3}$ absorption edges provides information on the matrix elements $F_{lm,l'm'}$ ($l, l' = 0$ or 2). This is the case for all experiments to date; that is, one can obtain information on the matrix elements $F_{lm,l'm'}$ with $|l - l'| = 0$ or 2 and one cannot in principal obtain information for $|l - l'| = 1$.

In this paper we investigate an experiment on inelastic x-ray scattering (IXS) which contains information on $F_{lm,l'm'}$ with $|l - l'| = 1$. We propose an experiment for performing on low- Z matter from which the matrix elements $F_{00,1m}$ can be easily extracted.

Recently a method for calculating the double-differential cross section of the inelastic x-ray scattering associated with low- Z K-level excitations of solids was developed [4, 5]. Both the first- and second-order contributions of perturbation theory were included. The first-order term proved to be determined by the p partial local densities of electron states (the p-LDOS) whereas the second-order term is determined by the s-LDOS. It was shown that by varying the initial photon energy one can significantly change the relative contributions of first- and second-order processes and therefore determine s- and p-LDOS. Our calculations of inelastic x-ray scattering fine structure for graphite crystal are in agreement with the available experimental spectra [6].

The expression for the double-differential cross section of the inelastic x-ray scattering in low- Z -atom crystals has the form [4]

$$\frac{d^2\sigma}{d\hbar\omega_1 d\Omega} = r_0^2 \frac{\omega_1}{\omega_0} (\mathbf{e}_0, \mathbf{e}_1)^2 S(\mathbf{q}, \omega_0) \quad (4)$$

where r_0 is the classical electron radius, ω_0 , ω_1 and \mathbf{e}_0 , \mathbf{e}_1 are frequencies and polarization vectors of incident and scattered radiation, respectively, and \mathbf{q} is the transferred wave vector. The expression for $S(\mathbf{q}, \omega_0)$ in the case of a low- Z -atom crystal with arbitrary symmetry has the form

$$S(\mathbf{q}, \omega_0) = \left(\sum_{\alpha} \sum_{\beta} F_{1\alpha,1\beta}(\varepsilon) q_{\alpha} q_{\beta} \right) K_1^2 + (F_{00,00}(\varepsilon)) |K_0|^2 + 2K_1 \text{Im}(K_0) \left(\sum_{\gamma} F_{00,1\gamma}(\varepsilon) q_{\gamma} \right) \quad (5)$$

where

$$K_0 = \frac{1}{3} \left[\frac{E - \varepsilon_0}{E - \varepsilon} \int R_0(r, \varepsilon) \frac{\partial U(r)}{\partial r} H_{E,1}^+(r_{>}) R_{E,1}(r_{<}) r' \varphi_0(r') r^2 dr r'^2 dr' + \frac{i \varepsilon - \varepsilon_0}{\hbar E - \varepsilon} \int R_0(r, \varepsilon) r \frac{\partial \varphi_0(r)}{\partial r} r^2 dr + \frac{E' + \varepsilon_0}{E' + \varepsilon} \int R_0(r, \varepsilon) \frac{\partial U(r)}{\partial r} H_{(-E'),1}^+(r_{>}) R_{(-E'),1}(r_{<}) r' \varphi_0(r') r^2 dr r'^2 dr' - \frac{i \varepsilon - \varepsilon_0}{\hbar E' + \varepsilon} \int R_0(r, \varepsilon) r \frac{\partial \varphi_0(r)}{\partial r} r^2 dr \right]$$

$$K_1 = \sqrt{\frac{1}{3}} \int R_1(r, \varepsilon) r \varphi_0(r) r^2 dr$$

$$\varepsilon = \hbar\omega_0 + \varepsilon_0 - \hbar\omega_1.$$

ε is the energy of the final electron, $E = \hbar\omega_0 + \varepsilon_0$, $E' = \hbar\omega_1 - \varepsilon_0$, φ_0 and ε_0 are the wave function and the energy of the 1s state, $U(r)$ is the atomic potential, and the functions $H_{E,l}^{\pm}$

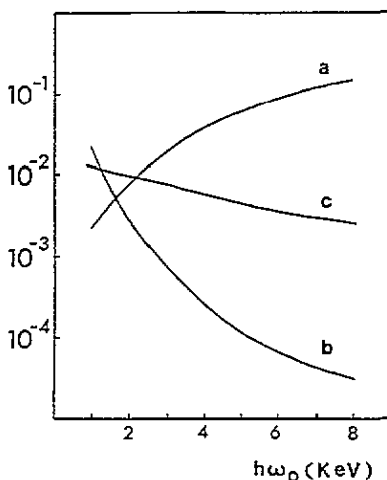


Figure 1. The coefficients $K_1^2 q_{max}^2$ (curve a), $|K_0|^2$ (curve b) and $2K_1 \text{Im}(K_0 q_{max})$ (curve c) in carbon versus the initial photon energy $\hbar\omega_0$ for energy loss $\hbar\omega_{loss}$ near the K edge of carbon. q_{max} is the value of the transferred wave vector for backscattering.

and $R_{E,l}$ are the solutions of the atomic radial Schrödinger equation which satisfy the same boundary conditions as the functions $h_{E,l}^+$ and $j_{E,l}$ respectively.

To estimate the value of each term in (5), we have calculated the coefficients $K_1^2 q_{max}^2$ (curve a in figure 1), $|K_0|^2$ (curve b in figure 1) and $2K_1 \text{Im}(K_0 q_{max})$ (curve c in figure 1) versus the initial photon energy $\hbar\omega_0$ for the energy loss $\hbar\omega_{loss} = \hbar\omega_0 - \hbar\omega_1$ near the K edge of carbon. Here q_{max} is the value of the maximal possible transferred wave vector. One can see from figure 1 that the double-differential cross section of inelastic x-ray scattering is determined by the s-LDOS for $\hbar\omega_0 < 2$ keV and small scattering angle. For $\hbar\omega_0 > 4$ keV the IXS is determined by the p-LDOS. In the middle region, $2 \text{ keV} < \hbar\omega_0 < 4 \text{ keV}$, the IXS is determined by both the s-LDOS and the p-LDOS. Moreover, in this case interference between matrix elements with different selection rules takes place. This provided us with the opportunity of extracting $F_{00,1\gamma}(\varepsilon)$ from inelastic scattering spectra. If two experimental spectra with transferred wave vectors q and $-q$ are obtained then the value $F_{00,1\gamma}(\varepsilon)$ can be easily extracted:

$$S(q, \omega_0) - S(-q, \omega_0) = 4K_1 \text{Im}(K_0) \left(\sum_{\gamma} F_{00,1\gamma}(\varepsilon) q_{\gamma} \right). \quad (6)$$

One can see from figure 1 that the most convenient conditions for extracting $F_{00,1\gamma}(\varepsilon)$ are found for $\hbar\omega_0 < 3$ keV.

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