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Theoretical study of focusing and double slit effects in x-ray photoelectron diffraction

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Abstract. Double-slit and forward focusing effects in XPD spectra from H2O and GeCl2 molecules have been theoretically studied within the single scattering approximation. In low- and intermediate-energy region, the double-slit effect can be observed. In contrast, this effect wears off and only forward focusing peaks with simple structure are given in high-energy region. The double-slit effects can be observed under special conditions where the two scattering amplitudes have finite overlap in the bisecting direction.

1. Introduction
X-ray photoelectron diffraction (XPD) is widely used as a tool of surface and near-surface structural analyses. The ejected electrons are strongly scattered in the forward direction which is called forward focusing effect. In XPD analyses direct structural information is obtained by using this effect [1]. This information is contrast to that from EXAFS. Besides the focusing effect, some interference can affect XPD pattern. Homonuclear diatomic molecules are often referred to as molecular double-slit. Cohen and Fano discussed this phenomenon by using a partial wave expansion within Born approximation [2]. Walter and Briggs used momentum representation and showed direct way to explain this phenomenon [3]. Recent experimental developments enable us to measure XPD patterns from fixed-in-space molecules and they provide us with useful information on photoemission dynamics [4, 5, 6]. With the help of these techniques, molecular double-slit experiments have been performed [7, 8]. In addition to diatomic models, another type of molecular double-slit can be considered; three-atom model, which contains one emitter A and two scatterers, B1 and B2. This model well describes Young’s double slit experiments. When $|AB_1|$ is equal to $|AB_2|$, a photoemission peak can appear in the direction of bisector of $\angle B_1AB_2$ in addition to the forward focusing peaks at $AB_1$ and $AB_2$ directions. So far no detailed study on this triatomic molecular double-slit effect has been undertaken except for [9]. In this paper, we show calculated angular distributions for photoelectrons from O 1s and Ge 1s core levels of H2O and GeCl2 to discuss the relative importance of the forward focusing and double-slit effects in XPD.
2. Theory
First principle many-body photoemission theory gives a XPD formula to study induced by x-ray photons with energy \( \omega \), in terms of the damped photoelectron wave function \( \psi_k^- \) under the influence of the optical potential [10, 11],

\[
I(k) = 2\pi|\langle \psi_k^- | \Delta | \phi_c \rangle|^2 \delta(E_0 + \omega - E_0^* - \epsilon_k),
\]

where \( k \) is the photoelectron momentum, \( \phi_c \) is the core wave function localized on the site A. The ground state energies with and without core hole are \( E_0^* \) and \( E_0 \), and \( \Delta \) is the electron-photon interaction operator. By use of site \( T \) matrix expansion, the amplitude is written as

\[
\langle \psi_k^- | \Delta | \phi_c \rangle = \langle \phi_{Ak}^- | \Delta | \phi_c \rangle + \sum_{\alpha(\neq A)} \langle \phi_{Ak}^- | t_\alpha g_A \Delta | \phi_c \rangle + \sum_{\beta \neq \alpha(\neq A)} \langle \phi_{Ak}^- | t_\beta g_0 t_\alpha g_A \Delta | \phi_c \rangle + \cdots,
\]

where \( \langle \phi_{Ak}^- \rangle \) is the decaying plane wave under the influence of the imaginary part of the optical potential, \( \langle \phi_{Ak}^- \rangle \) is the photoelectron wave function emitted from an x-ray absorbing atom A, \( g_0 \) is the decaying free Green’s function and \( g_A \) is the Green’s function influenced by the potential only on absorbing atom A. The first term in equation (2) describes the direct photoemission amplitude \( (Z_1) \) without scatterings from surrounding atoms. The second term describes the single scattering amplitude \( (Z_2) \), and the third term is the double scattering amplitude \( (Z_3) \). By applying multiple scattering renormalization, we obtain a full multiple scattering formula [12, 13]:

\[
\langle \psi_k^- | \Delta | \phi_c \rangle_\infty = \sum_{\alpha} e^{-ik R_\alpha} \sum_{LL'} Y_{L'}(\hat{k}) \left[ (1 - X)^{-1} \right]_{L'L} M_{LL',c},
\]

\[
(1 - x)^{-1} = 1 + X + X^2 + \cdots, \quad X^{\alpha \beta}_{LL'} = t_{L'}(k) G_{LL'}(k R_{\alpha \beta})(1 - \delta^{\alpha \beta})
\]

where the matrix \( X \) is labeled by atomic site \( \alpha, \beta, \ldots \) and angular momentum \( L = (l, m) \). \( R_\alpha \) is the position vector of scatterer \( \alpha \) measured from the x-ray absorbing atom A. The full multiple scattering is taken into account by use of the inverse matrix \( (1 - X)^{-1} \).

3. Results and discussion
In this section we discuss double-slit and focusing effects in photoelectron angular distribution from H2O and GeCl2. Geometrical setup is shown in figure 1. We use two linear x-ray polarizations, e (polarization vector) \( \parallel z \) and \( \parallel x \). The bond length of H2O is 0.958 Å and \( \angle \)HOH=104.5°, and the bond length of GeCl2 is 2.183 Å and \( \angle \)ClGeCl=100.3°. In the phase shift calculations, we use sufficiently accurate non-local optical atomic potential which takes important many-body effects into account [14]. To obtain physical insight we discuss the XPD patterns in the single scattering approximation,

\[
|Z_1 + Z_2|^2 = |Z_1|^2 + 2 \text{Re}(Z_1^* Z_2) + |Z_2|^2.
\]

The term \( |Z_2|^2 \) partly includes the Young’s double-slit effects because the interference of two photoelectron waves \( R_1 \rightarrow k \) and \( R_2 \rightarrow k \) correspond to the double-slit effects. The second term \( 2 \text{Re}(Z_1^* Z_2) \) describes the interference between the direct and the single scattering waves, and it partly includes the double-slit effect because \( Z_2 \) has the above two different path. \( 2 \text{Re}(Z_1^* Z_2) \) cannot be compared with classical double-slit because the direct amplitude \( Z_1 \) is characteristic of photoemission.
First we study in the case of linear polarization $e \parallel z$. Angular distributions of $|Z_2|^2$ for O 1s photoelectrons from H$_2$O are shown in figure 2. We observe prominent double-slit peaks in the direction of $z$ in low-energy region ($\epsilon_k \leq 300$ eV), whereas a small peak is observed at $\epsilon_k = 500$ eV. This behavior is explained by the sharp forward peak in the differential cross section (DCS) from a H atom, as shown in figure 3. In the direction of $z$, the scattering angle is about 50°, and the DCS is nearly zero at $\epsilon_k = 500$ eV. The ratio of $|Z_2|^2/|Z_1|^2$ is about 1/20 even at $\epsilon_k = 100$ eV, because H atom is a weak electron scatterer. It is thus difficult to observe the double slit effects in $|Z_1 + Z_2|^2$. Yamazaki et al. measured angular distributions of O 1s photoelectrons from H$_2$O at $\epsilon_k = 14$ eV [9]. They numerically studied the contributions of the three terms in equation (4) to the XPD pattern and found that the double-slit interference is negligible.

In contrast to H$_2$O, GeCl$_2$ gives no double-slit peak for the observed bond angle (100.3°) at $\epsilon_k = 100$ eV (figure 4 (a)). Figure 5 shows the DCS from a Cl atom at 100 eV, which oscillates as a function of scattering angle; this is known as Ramsauer-Townsend effect [15]. At 50° the DCS happens to be zero. We thus expect to observe no double-slit effect. When we hypothetically set ClGeCl at 60°, a double-slit peak appears (see figure 4 (b)), because the DCS at 30° is finite. Figure 6 separately shows the three terms in equation (4) and the total intensity at ClGeCl=60°. In this case, $|Z_2|^2$ has appreciable intensity, but $2\text{Re}(Z_1^*Z_2)$ cancels the slit peak in $|Z_2|^2$. In high kinetic energy region ($\epsilon_k \sim 1000$ eV), $|Z_2|^2$ does not show double-slit pattern even when ClGeCl=60°, and two focusing peaks with simple structure are observed.

When the x-ray polarization is parallel to the $x$ axis ($e \parallel x$, see figure 1), the XPD intensity vanishes in the direction parallel to $z$ axis. In the plane-wave expression, the amplitude $Z_2$ is
Figure 5. The differential cross sections from Cl atom at 100 eV, which is normalized so that the intensity at 0° are unity.

Figure 6. Ge 1s XPD pattern from GeCl₂ (\(\epsilon_k = 100\) eV). We set \(\angle\text{ClGeCl}= 60°\). Polarization vector is parallel to the \(C_2\) axes.

Explicitly given for the photoionization from a deep s orbital,

\[
Z_2 \propto e^{i\hat{\alpha}1} \rho(1) \sum_\alpha \frac{\cos \hat{\theta}_\alpha f_\alpha(\theta_\alpha)}{R_\alpha} e^{ikR_\alpha(1-\cos \theta_\alpha)} , \quad \cos \hat{\theta}_\alpha = e \cdot \hat{R}_\alpha \tag{5}
\]

where the radial dipole integral \(\rho(1)\) weakly depends on \(\epsilon_k\), and \(\theta_\alpha\) is the scattering angle. In the case of \(\mathbf{k} \parallel z\), we notice that \(\cos \hat{\theta}_1 = -\cos \hat{\theta}_2\) for the symmetric arrangement considered here. Then the coherent sum of two single scattering amplitudes in equation (5) vanishes at the direction parallel to \(z\) axis. The factor \(\cos \hat{\theta}_\alpha \propto Y_{10}(\hat{R}_\alpha)\) comes from \(G_{L,0}\) in equation (3), which exists in all power of \(X\). Thus the XPD intensity vanishes along \(z\) axis, even if the spherical wave effect and the full multiple scatterings are taken into account.

4. Concluding remarks

At low and intermediate energies the double-slit effects have appreciable contribution only when DCS is non-negligible at the bisecting angle. We should note that the interference term \(2\Re(Z_1 Z_2^\ast)\) can make negative contribution at the expected direction and can cancel the double-slit effects. For the systems considered here, the double-slit effects have small influence on XPD patterns.

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