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Original Paper

Theoretical Study of Plasmon Loss Peaks in Core-level Photoemission Spectra: Energy and Angular Dependence

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In the present paper surface and bulk plasmon loss features excited by high-energy photons are studied. In particular the relative importance of the interference is investigated in detail. Although the interference terms should drop out as a function of the photoelectron energy, we find those terms decays very slowly and are still quite important even at photon energy 5 keV. Those for the surface plasmon losses do not show drop out even in this quite high-energy region. The extrinsic losses play the most important role among the three terms. In the low-energy region, the loss features show the angular dependence in contrast to those in the high-energy region.

1. Introduction

The appearance and the origin of plasmon loss satellites in core-level photoemission spectra has extensively been studied, however some problems such as relative importance of “intrinsic” and “extrinsic” losses are still unsolved. In the three-step model of photoemission, the plasmon excitation process can be classified into intrinsic and extrinsic ones. The plasmon excitation processes by the core-hole potential are called intrinsic, which are contained in the one-electron spectral function. Inelastic plasmon losses during photoelectron transport to the surface are called extrinsic. In the one-step model of the photoemission, we can expect the quantum mechanical interference between these two processes [1-5]. This interference effect, which becomes very important when the kinetic energy of the photoelectron is small [6], has been often neglected in the analyses of experimental data. These processes can be distinguished by the measurement of the angular dependence of the plasmon satellite peaks. For example, the loss peaks will be of purely extrinsic origin when measured at forbidden direction if we neglect the photoelectron diffraction effects.

In the previous paper we have only shown the calculated results for the two different photon energies 125 and 1486.6 eV. There the experimental results are also shown which are compared with the calculated results: That quantum calculations well explain the experimental features [5].

We should also refer to the alternative semi-classical dielectric response model for the same problem [7], which is compared with recent experimental result [8]. These semi-classical approaches require much less computation cost, and also are even quantitative.

In the present paper plasmon loss features excited by high-energy photons are studied. In particular the relative importance of the interference is investigated in detail. As pointed out by Hedin [9] the interference terms should drop out as a function of the photoelectron energy. We have an interest in the question how fast they drop out.

We perform a full-quantum-mechanical calculation taking the interference into account by use of the optical potential, which provides useful information on the depth, energy and angular dependence of the loss peaks.

2. Theory

For the present numerical calculations we apply the model used in ref. [4]. We considered a half-infinite electron gas with an embedded ion at distance $z_c > 0$ from the surface. We take the self energy as zero outside the solid, and $-i\Gamma$ inside. Neglecting the influence of the embedded ion, the fluctuation potential can be written

$$V^{\boldsymbol{q}}(\boldsymbol{r}) = \exp(i\boldsymbol{Q}\cdot\boldsymbol{R})V^{q}(z),$$
$$\boldsymbol{r} = (\boldsymbol{R}, z), \qquad \boldsymbol{q} = (\boldsymbol{Q}, q).$$

For the photoelectron state we use the time-inverted LEED state $|\tilde{\boldsymbol{k}}\rangle$, where $\tilde{\boldsymbol{k}}$ is related to the photoelectron momentum $\boldsymbol{k}$

$$\tilde{\boldsymbol{k}} = \sqrt{k^2 + 2(\phi + i\Gamma)}$$

where ϕ is the work function (ϕ > 0). Taking the core function as having zero extent, we obtain the simple formula for the photoelectron current $J_{\boldsymbol{k}}(\omega; z_c)$ from many-body scattering theory [4]. We measure the current density of photoelectrons with momentum $\boldsymbol{k}$ excited by X-ray photons with energy ω

$$J_{\boldsymbol{k}}(\omega; z_c) \sim -\frac{\left|\langle\tilde{\boldsymbol{k}}|\Delta|c\rangle\right|^2}{\pi} e^{-2z_c \mathrm{Im}\tilde{k}} \times \sum_{\boldsymbol{q}} \int f(z) f(z')^* \mathrm{Im} W(\boldsymbol{q}, z, z'; \omega - \varepsilon_k) dz dz', \qquad (1)$$

$$f(z) = \frac{\delta(z - z_c)}{\omega - \varepsilon_k} + \frac{i}{\kappa} e^{i(\tilde{k}-\kappa)(z-z_c)}\theta(z_c - z), \qquad (2)$$
$$\kappa = \sqrt{2(\omega - \phi + i\Gamma) - |\boldsymbol{Q} + \boldsymbol{\kappa}|^2},$$

where $\left|\langle\tilde{\boldsymbol{k}}|\Delta|c\rangle\right|^2$ describes the direct photoemission intensity excited from a core c on the site z_c: Δ is the electron-photon interaction operator, $|c\rangle$ is the core function.The first term of $f(z)$ is related to the intrinsic amplitude, and the second is related to the extrinsic amplitude. In this theoretical approach, the screened Coulomb interaction W plays a crucial role to describe the plasmon loss features. We should notice that this theory cannot take into account the elastic scatterings from surrounding atoms.

For practical purposes we have to introduce some approximation for W near solid surfaces. Here we use the screened Coulomb potential proposed by Bechstedt *et al.* [10].

Experimental photoemission intensity is obtained by the sum over lattice sites to get convergence

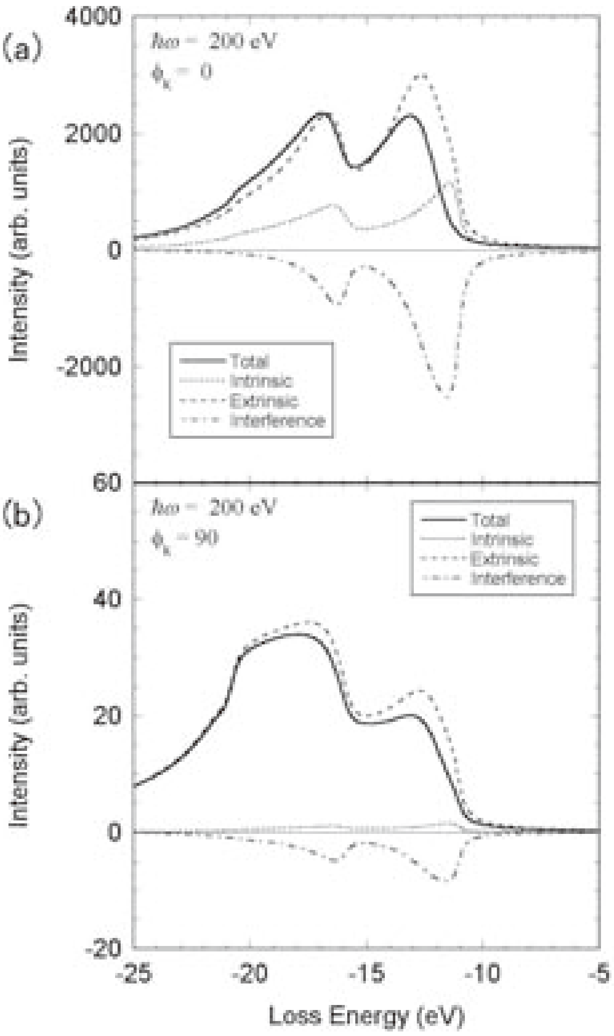

Figure 1: Calculated Al $2p$ plasmon loss structures excited by photon energy $\hbar\omega$ = 200 eV at ϕ_k =0° (a) and 90° (b). Energy is measured from the Al $2p$ elastic peak.

$$J_{\boldsymbol{k}}(\omega) = \sum_{z_c} J_{\boldsymbol{k}}(\omega; z_c). \qquad (3)$$

In his final article, Hedin has pointed out that the interference terms should drop out for the bulk plasmon losses in the high-energy limit [9]. We, however, have no clear-cut information on whether they can be neglected or not in our typical “high-energy” region, for example, 3 - 7 keV.

3. Calculated results

In the previous paper, Uwatoko *et al.* have studied the plasmon loss peaks of Al $2p$ at intermediate energies [5], which successfully explain the excitation energy and angular dependence of the loss peaks: Destructive interference plays an important role especially at low energy

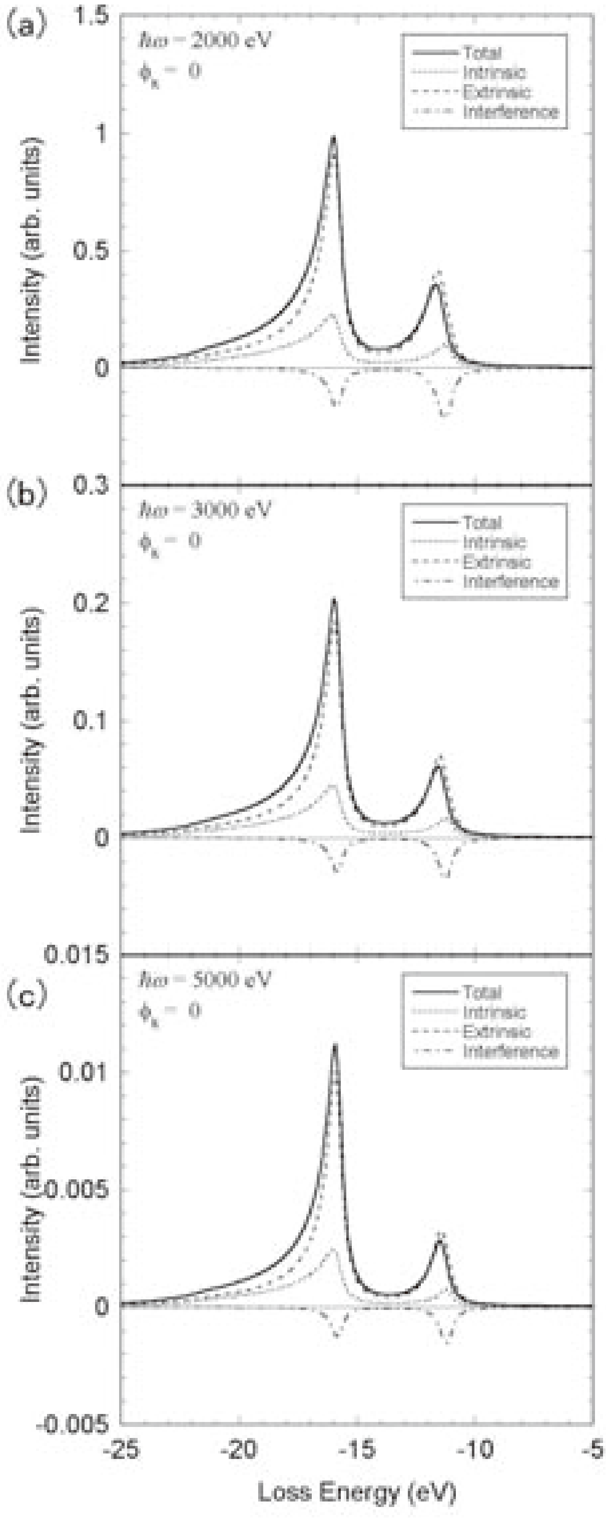

Figure 2: Calculated Al $2p$ plasmon loss structures at ϕ_k =0º excited by photon energy $\hbar\omega$ = 2000 (a) 3000 (b) and 5000 eV (c). Energy is measured from the Al $2p$ elastic peak.

$\hbar\omega$ = 125 eV, whereas extrinsic contribution is dominant at higher energy $\hbar\omega$ = 1486.6 eV.

We now show some new calculated results on the basis of eqs.(1) and (2). Parameters used in the present calculations are the same as those used in the previous paper [5]. We assume that the X-ray polarization is parallel to the x-axis, and we measure the photoelectron current intensity $J_{\boldsymbol{k}}(\omega)$ at the take-off angle 60º from the Al (110) surface.

Before we discuss the loss features in the high-energy region, the calculated spectra at $\hbar\omega$ = 200 eV (ε_k = 128 eV) are shown for comparison. Figure 1 (a) and (b) show the calculated loss spectra measured at ϕ_k = 0º and 90º. Three different contributions, “extrinsic”, “intrinsic” losses and their “interference”, are separately shown. Their sum, “total”, is also shown. We observe two peaks associated with surface and bulk plasmon losses. The direct photoemission intensity show a prominent maximum at ϕ_k = 0º and minimum at ϕ_k =90º. In particular photoemission from the deep s orbital, the intensity should perfectly be suppressed as far as we neglect elastic scatterings from surrounding atoms. The loss intensity in Fig 1 (b) is thus much smaller than that in Fig. 1 (a). In Fig. 1 (b) the relative contribution of the intrinsic loss to the extrinsic loss is very small compared with that in Fig. 1 (a). When we neglect the elastic scatterings, the intrinsic loss processes provide no chance to change the photoelectron propagation. On the other hand, the extrinsic loss processes provide a chance to change the propagation during the inelastic scatterings. Considerable contribution of large angle plasmon loss scatterings at low energy can give different loss features for different $\hat{\boldsymbol{k}}$. In this energy region (small mean-free-path), the spectra are very sensitive to the surface, which is reflected in the intensity ratio of the surface plasmon loss to that of bulk loss: In Fig. 1 (a) both have nearly the same intensities.

Next we discuss the loss features in the high-energy region 2 keV $\leq \hbar\omega \leq$ 5 keV. Figure 2 (a), (b) and (c) show the calculated loss spectra at $\hbar\omega$ = 2, 3 and 5 keV measured at ϕ_k = 0º. We observe quite different features from those in Fig. 1 and also in ref. [5]. Here we observe distinct surface and bulk plasmon loss peaks, and very weak energy dependence. Another interesting result is that the interference terms do not drop out even in this quite high-energy region, $\hbar\omega$ = 2 - 5 keV. The rate of the drop out relative to the total intensity looks different for bulk and surface plasmon losses: In the former the rate is fast compared with that in the latter.

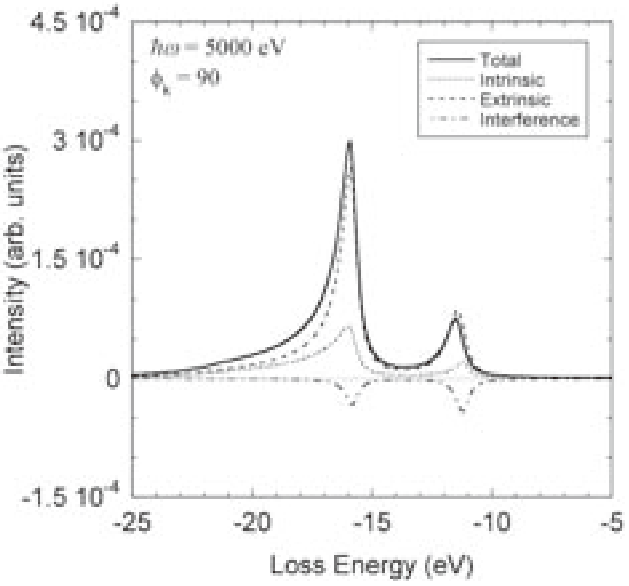

Figure 3: The same as Fig. 2 but at ϕ_k = 90º and $\hbar\omega$ = 5000 eV. Energy is measured from the Al $2p$ elastic peak.

Hedin's prediction that the interference terms should decay is obtained for the bulk plasmon loss. We have no idea about the drop out for surface plasmon losses.

Figure 3 shows the loss spectra excited by X-ray photons with energy 5 keV measured at ϕ_k = 90º. In contrast to the low energy excitation, the spectral feature is quite similar to that in Fig. 2 (c) except their overall intensities: The intensity in Fig. 3 is much smaller than that in Fig. 2 (c) because of the factor $|\langle\tilde{\boldsymbol{k}}|\Delta|c\rangle|^2$. From these results the plasmon loss features in the high-energy region (> 1 keV) only depend on ε_k not on $\boldsymbol{k}$.

4. Concluding Remarks

In this study we calculate the single plasmon (surface + bulk) loss structures of Al $2p$ photoemission excited by high energy X-rays. The extrinsic term is dominant in the energy region $\hbar\omega$ > 1 keV over the intrinsic and the interference terms. The interference term is not negligibly small even at 5 keV. This result may sound funny because the intrinsic approximation is equivalent to the sudden approximation. In the high-energy region the core-hole is "suddenly" produced. Intrinsic losses, however, are not predominant even at these high-energies. Near the plasmon threshold, the loss features are lost in the background. This can be clearly related to "adiabatic" core-hole production.

The interference terms for the bulk plasmons drop out faster than those for the surface plasmons, but the rate is still quite slow.

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