

## The role of the electron inelastic scattering processes in EELFS spectra

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The pseudopotential model is used for the evaluation of the core level ionisation intensities (by the electron impact). The central atom effective phase shift is calculated for K EELFS spectra in this framework. The Si K EELFS spectra were calculated and compared with the experimental data. The comparison of experimental and calculated results is performed on the base of the electron dispersion law which takes into account electron-plasmon interaction.

**Keywords:** EELFS, EXAFS, Orthogonalized Plane Wave (OPW) approximation

### 1. Introduction

Extended energy loss fine structures (EELFS) are located above core - level ionisation thresholds in electron energy loss spectra. EELFS are formed by secondary electron coherent scattering, i.e. EELFS are EXAFS - like oscillations (De Crescenzi, 1995). However, the EELFS spectrum is created as a result of the core level ionisation by the electron impact (Fig. 1). Accordingly the oscillating part of EELFS spectrum in the single scattering approximation has the form (Guy et al., 2000):

$$\chi(k, R_j) = \sum_{j \neq 0} \text{Re} \left[ \mu_j \exp(i2\delta^0) W_j(k) f_j(k, \pi) \exp(-2R_j/\lambda) \frac{\exp(i2kR_j)}{ikR_j^2} \right], \quad (1)$$

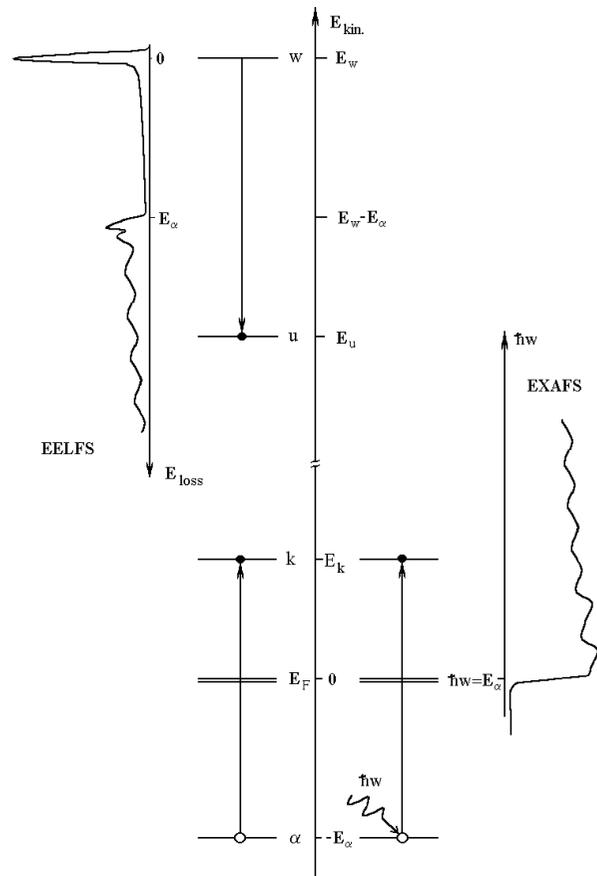
where  $R_j$  is the distance between ionised (0-th) and neighbouring ( $j$ -th) atoms,  $k$  is the wave number of the secondary electron,  $f_j(k, \pi)$  is the back-scattering amplitude,  $\lambda$  is the secondary electron mean free path, and  $W_j(k)$  is the Debye - Waller factor. In this process the energy conservation law implies that:  $E_w - E_u = E_k + E_\alpha$ , where  $E_w$  is the incident electron energy,  $E_u$  is the energy of an inelastically scattered electron recorded in the experiment,  $E_k$  is the secondary electron energy and  $E_\alpha$  is the core level electron binding energy. In Eq. (1)  $2\delta^0$  is the effective phase shift on the 0-th atom which depends on partial phase shifts and the amplitude of core level ionisation by the electron impact and is determined by:

$$|\mu| \exp(i2\delta^0) = \sum_l \mu_l (-1)^l \exp(i2\delta_l^0), \quad (2)$$

where  $\mu_l$  are the coefficients of decomposition in Legendre series of the function:

$$\frac{\langle T^*(\vec{k}) T(\vec{k}') \rangle_{\vec{k}}}{\langle T^*(\vec{k}) T(\vec{k}) \rangle_{\vec{k}}} = \sum_l \mu_l P_l \left[ \cos \left( \vec{k}, \hat{\vec{k}} \right) \right], \quad (3)$$

where  $|\vec{k}| = |\vec{k}'|$ ,  $T(\vec{k})$  ( $T(\vec{k}')$ ) is the secondary electron creation amplitude with a wave vector  $\vec{k}$  ( $\vec{k}'$ ). Note that in the case of the core level radiative ionisation (as in EXAFS) in Eq. (3) only  $\mu_{l=1}$  is not equal to zero, and the Eq. (1) becomes the usual EXAFS formula.



**Figure 1**  
Diagrams of electron transitions forming EELFS and EXAFS spectra

### 2. Evaluations in ortogonalized plane wave approximation

In the ortogonalized plane wave (OPW) approximation and in the case of the core level ionisation by electron impact, the transition amplitude in Eq. (3) may be written in the form:

$$T(\vec{k}) = 4\pi \left[ \frac{1}{\mathbf{x}^2} - \frac{1}{w^2} \right] \left\langle k \left| \exp(i\vec{\mathbf{x}}\vec{r}) - \xi \right| \alpha \right\rangle, \quad (4)$$

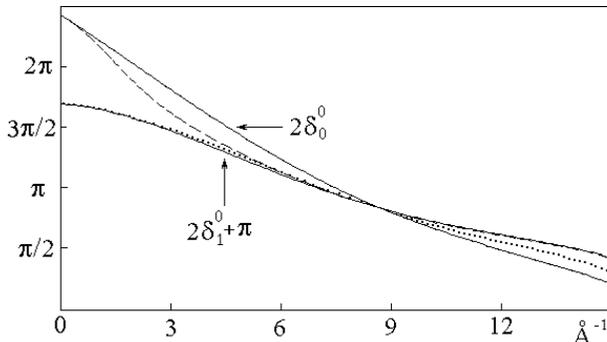
where  $\vec{\mathbf{x}} = \vec{w} - \vec{u}$  is the transferred momentum,  $\vec{w}$  is the initial electron wave-vector and  $\vec{u}$  is the inelastically scattered electron wave vector. The core level state is denoted as  $|\alpha\rangle$  and the secondary electron state -  $|k\rangle$ . In Eq. (4) the correlation term  $\xi$  is a consequence of the OPW approximation for secondary electron wave functions. Note that the influence of the correlation term is larger in multiplet decomposition of  $T(\vec{k})$ , i.e. in determining the effective phase shift (Eq. (2)). In the correct approach (OPW), the correlation term should be determined by:  $\xi = \sum_{|\alpha'\rangle} \langle \alpha' | \exp(i\vec{\mathbf{x}}\vec{r}) | \alpha \rangle$ , where  $|\alpha'\rangle$  is one of all the atomic core states. Unfortunately this approach is difficult in real practice. As a rule, the correlation term is chosen as:  $\xi = \langle \alpha | \exp(i\vec{\mathbf{x}}\vec{r}) | \alpha \rangle$  (Fujikawa et al., 1988). This is a reasonable approximation for  $\mathbf{x}^2/\alpha^2 \ll 1$  (here  $\alpha$  is the inverse radius of localisation of a core level electron wave function), i.e. in the experiments with the incident electron forward scattering geometry and in the case when the incident electron energy is larger than the

core level electron binding energy. In alternative situations, for example, in the experiments with the incident electron back scattering geometry such a simple approximation is not correct.

In this paper for the 1s core level excitation process the correlation term was simulated on the basis of evaluations of monopole and dipole transition intensities. Evaluations of these transition intensities can be carried out for the hydrogen atom (Saldin & Yao, 1990), where the Coulomb function is chosen as the secondary electron wave function. In  $\kappa^2/\alpha^2 \ll 1$  asymptotic evaluations of monopole and dipole transition intensities (Saldin & Yao, 1990) give that the dipole transition intensity is larger when  $\kappa^2/\alpha^2 \ll 1$  and  $p^2/\kappa^2 \geq 1$  and the monopole transition intensity is larger when  $\kappa^2/\alpha^2 \ll 1$  and  $p^2/\kappa^2 < 1$ . Evaluations of monopole and dipole transition intensities have been carried out in this approach for  $p^2/\alpha^2 \ll 1$ . As a result the monopole transition dominates for  $p^2/\alpha^2 \ll 1$  and  $\kappa^2/\alpha^2 \ll 1$  and the dipole transition dominates for  $p^2/\alpha^2 \ll 1$  and  $\alpha^2/\kappa^2 \ll 1$ . This asymptotic behaviour may be taken into account in the pseudopotential approach if the correlation term is chosen as  $\xi = \left(4^n \alpha^2 / (4^n \alpha^2 + \kappa^2)\right)^2$ , where  $n = \alpha^2 / (\alpha^2 + \kappa^2)$ .

### 3. Application for Si K - EELFS spectra

Let us examine the effect of this result on the real EELFS spectra. The main result of the effect of non-dipole processes on the EELFS spectra is the appearance of the central atom middle effective phase shift in Eq. (1). Silicon effective phase shifts for forward and back inelastic scattering of an incident electron are shown in Fig. 2 in comparison with partial phase shifts (FEFF7). The calculation was carried out at  $E_w = 100$  keV ( $E_\alpha = 1839$  eV).

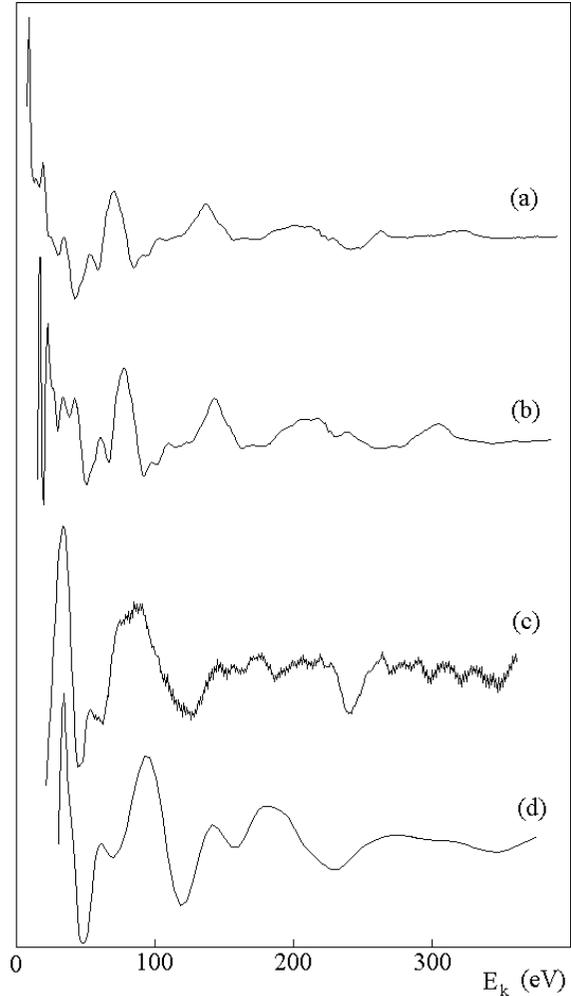


**Figure 2**  
The comparison of effective phase shifts calculated for forward (dashed line) and back (point line) inelastically scattering of the incident electron with partial ( $l = 0$  and  $l = 1$ ) phase shifts

This high value of  $E_w$  was chosen for the most clarity. The behaviour of the central atom effective phase shift essentially depends on  $E_w$  and differs from partial phase shifts. At small values of the secondary electron wave number the behaviour obtained is a direct consequence of choosing the correlation term, i.e. of choosing the asymptotic.

Along with the problem of the effect of non-dipole processes on the EELFS spectra there is one more problem which is general for all EXAFS - like phenomena. Namely, it is the problem of the secondary electron dispersion law. Inelastically scattering processes of the secondary electron determine the deviation of the secondary electron dispersion law from the free electron dispersion law. The secondary electron dispersion law is determined from the equation:

$E_k - \frac{\hbar^2}{2m} k^2 - \text{Re}\Sigma(E_k, k) = 0$ . Taking into account the free electron gas plasmon excitation processes gives the self-energy part in the form (Grebennikov, 2000):  $\Sigma(E_k, k) = -\frac{i\hbar w_p}{2ka_0} \ln \frac{k_0 + k}{k_0 - k}$ , where  $k_0 = \sqrt{2m(E_k - \hbar w_p)/\hbar}$  and  $\hbar w_p$  is the plasmon energy.



**Figure 3**  
The comparison of calculated (FEFF7) (a) and experimental (b) K - EXAFS spectra of Si with calculated (d) and experimental (c) Si K - EELFS spectra

Experimental K - EELFS spectra of Si were measured using an Auger microprobe JAMP - 10S in the incident electron back-scattering geometry at the incident electron energy of 3 keV. The corresponding calculation was carried out on the base of Eq. (1) using the crystallographic parameters of local atomic structure and secondary electron scattering parameters calculated by FEFF7 (Rehr et al., 1992). The angle averaged effective phase-shift of an inelastically scattering incident electron was calculated at the incident electron energy corresponding to the experimental one. The secondary electron dispersion law was simulated using as a test the experimental Si EXAFS spectrum and the calculated (FEFF7) one (Fig. 3 (a, b)), where the resonance approximation was used for the

self-energy part  $\left(\Sigma\left(E_k = \frac{\hbar^2}{2m}k^2, k\right)\right)$  and  $\hbar\omega_p = 25$  eV. Using

the same dispersion law the comparison of the experimental EELFS spectrum and calculated one is shown in Fig. 3 (c, d).

The comparison of EELFS and EXAFS spectra (Fig. 3) shows their differences that is the result of the non-dipole transition effect on EELFS spectra formation. The pseudopotential for the calculation of the core level ionisation (by the electron impact) intensities was suggested. A satisfactory agreement between experimental and calculated Si K EELFS spectra was obtained in this approach. This demonstrates the importance of taking

into account the non-dipole contributions in EELFS spectra as suggested with the present approach.

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### References

- De Crescenzi, M. (1995). *Surf. Sci. Rep.* **12**(3/4), 91 - 175.  
Fujikawa, T., Takaton, S., & Usami, S. (1988). *Jap. J. Appl. Phys.* **27**(3), 348 - 353.  
Grebennikov, V. I. (2000). *Fizika Met. i Metalloved.*(Russian) **89**(25), 5 - 9.  
Guy, D. E., Grebennikov, V. I. & Ruts, Yu. V. (2000). *J. Struct. Chem.* (Russian) **41**(6), 1267 - 1275  
Rehr, J.J., Albers, R.C., & Zabinsky, S.I. (1992). *Phys. Rev. Lett.* **69**, 3397 - 3400  
Saldin, D. K. & Yao, J. M. (1990). *Phys. Rev. B* **41**(1), 52 - 61.