Calculation of relativistic differential cross-sections for use in microanalysis

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To obtain quantitative information about specimens from experimental methods like elemental mapping or electron energy loss spectroscopy (EELS) one needs an accurate knowledge of the appropriate cross-sections. Using EELS it is possible to determine the chemical composition of the specimen quantitatively, if the double differential cross-section is known.

Modern transmission electron microscopes (TEM) operate at acceleration voltages of several hundred kV. Incident electrons with energies about 200 keV have velocities of $2.08 \cdot 10^8$ m/s corresponding to 70% of the speed of light. It is obvious that retardation, the delay of the interaction due to the finite velocity of light, is not negligible for these velocities. Therefore it is indispensable to account for relativistic effects in the calculations for such energies.

Surprisingly standard software packages often use programs like Egertons SigmaK [1] that are based on "kinematically corrected" and not on fully relativistic expressions for the cross-section. Here "kinematically corrected" means that the relativistic expressions for energy and momentum are inserted in non-relativistic equations. Doing this, the influence of retardation and the magnetic interaction between the incident and the atomic electron is completely neglected. Knippelmeyer et al. [2] have shown that in fully relativistic calculations the differential cross-section differs significantly from the one in SigmaK. Figure (1) shows the angular distribution for relativistic and non-relativistic calculations of the cross-sections using the dipole approximation [3]. We find that the difference between relativistic and non-relativistic calculations is not only quantitative but also qualitative, even for acceleration voltages of 300 kV.

Therefore we calculate the double differential cross-section in central-field approximation using relativistic equations. In our model we assume only single scattering, which restricts us to specimens thinner than the mean free path length of the incident electron. We neglect the influence from neighbouring atoms, by using the atomic model for the scattering process. Due to the large energy difference between the ionised and the scattered electron, exchange effects can be neglected. In our model we use single particle wave functions. The use of Feynman's propagator theory, Coulomb gauge and first order Born approximation leads to the equation

$$\frac{d\sigma}{d\Omega dE} = \alpha^2 \frac{4\,k_i^2}{\beta_i^2} \frac{k_i}{k_f} \frac{1}{2} \sum_{m,n} \left\{ \frac{1}{q^4} \left| \langle m | e^{i\vec{q}\vec{r}} | n \rangle \right|^2 + \left(\frac{\vec{\beta}_i \vec{t}}{q^2 - \left(\frac{\Delta E}{\hbar c}\right)^2} \right)^2 \left| \langle m | \vec{\alpha} \vec{t} e^{i\vec{q}\vec{r}} | n \rangle \right|^2 \right\}.$$

Here is $\alpha \approx 1/137$ the fine structure constant, $\vec{\beta}_i$ the velocity of the incident particle normalised to the vacuum speed of light, $\vec{r}$ the position of the atomic electron, $\vec{\alpha}$ the vector of Dirac-matices, $\vec{k}_i$, $\vec{k}_f$ the wave vectors of the incident and the scattered electron, $\vec{q}$ the momentum transfer, $\vec{t}$ an unit vector perpendicular to $\vec{q}$ and situated in the scattering plane, $|n\rangle$ the initial and $|m\rangle$ the final state of the atom and ΔE the energy loss of the incident electron. The indices m and n of the sum stand for quantum numbers of the relevant states. The factor $4k_i^2/\beta_i^2$ arises from the kinematics of the scattering process. In the non-relativistic case it reduces to $4m_0^2 c^2/\hbar$. The first term contains the Coulomb interaction between the incident and the atomic electron and in the second retardation effects are considered. The first term is equal to the "kinematically corrected" calculations. Therefore the differences between the relativistic and the nonrelativistic values for the cross-sections are given by the second term. For light elements ($(Z\alpha)^2 << 1$) or loosely bound states it is possible to use Darwin instead of Dirac wave functions. The Darwin wave function contains a scale factor, a bispinorial factor and a Schrödinger wave function. In this approximation the matrix-elements can be written as integrals of well-known Schrödinger wave functions, which are a product of a radial and an angular function. In the hydrogenic model all integrals can be solved analytically [2]. These calculations are however limited to K-shell ionisation. Our approach describes higher shell ionisation as well and results in an analytically solvable angular part and in radial integrals, which must be computed numerically. The resulting expressions can be viewed as an extension of the non-relativistic formulae [4]. We calculate the initial radial wave functions as a self-consistent solution of the Schrödinger equation, using a slightly modified program written by Hamann [5]. As final state, a continuum state is calculated by solving the Schrödinger equation with fixed energy. The continuum radial functions can be

normalized to a finite energy interval [6]. The radial integrals have the form of a Hankel-transform, which consist of a function with the radius as argument multiplied by a Bessel-function with the radius r and the momentum transfer q in the argument. Defined on a logarithmic mesh, these integrals can be recognized as a convolution and solved using fast Fourier transforms. We used a program, written to compute such expressions, based on Talmans routine [7] and modified by A. Weickenmeier for the calculation of the Bessel-function for orders greater than one.

The resulting program delivers the relativistic cross-sections for arbitrary shell ionization. In these calculations relativistic effects which lead to even qualitative difference like figure (1) shows are considered. Using these cross-sections one should obtain more accurate results in microanalysis.

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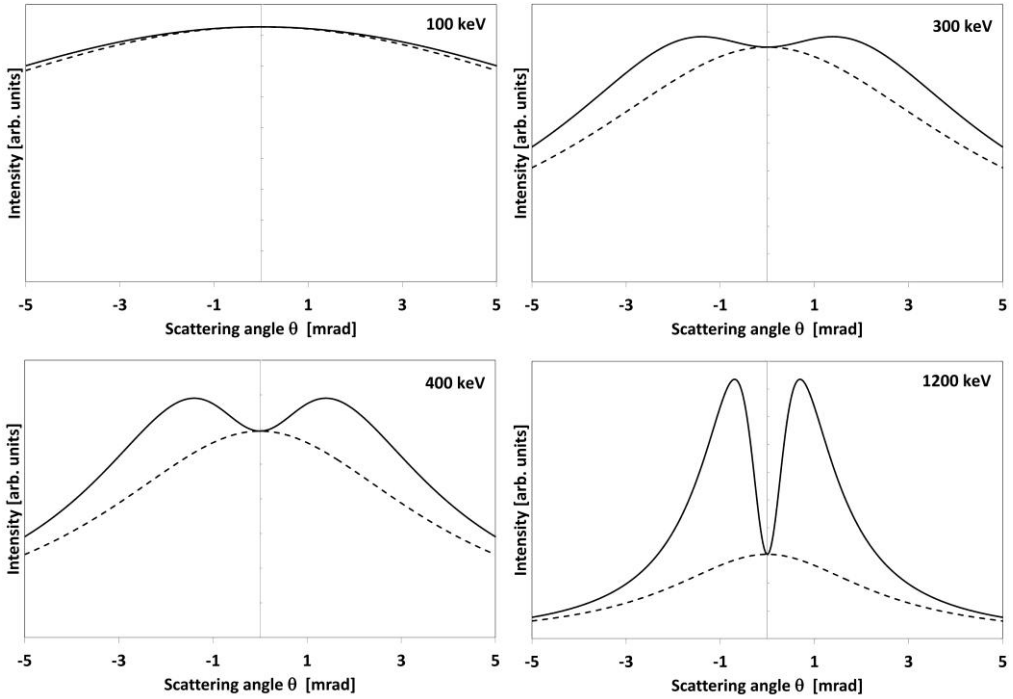

Figure 1. Comparison of relativistic (solid line) and non-relativistic (dashed line) angular distribution $f(\beta, \theta/\theta_E)$ for different acceleration energies at silicon K-loss (1839 eV). The functions are computed due to Fanos equations [3].

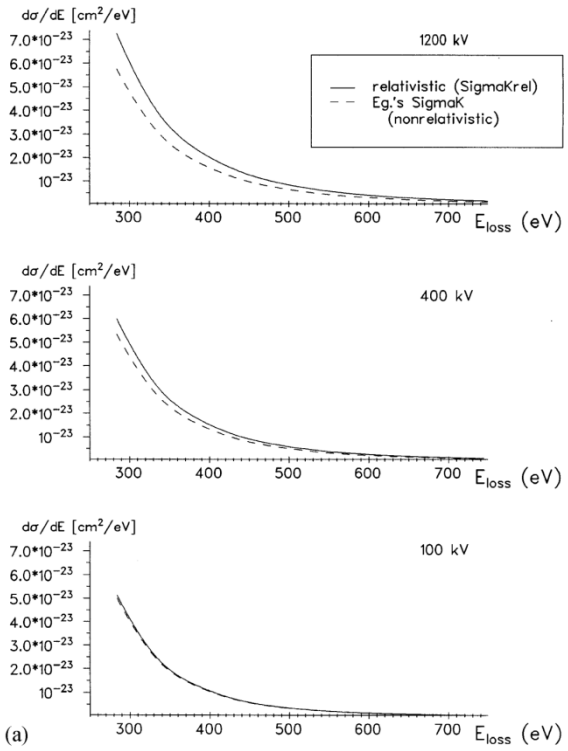

Figure 1. Comparison of energy-differential cross-sections calculated fully relativistically by Knippelmeyer's SigmaKrel [2] (solid line) and with Egerton's SigmaK [1] in the case of carbon at varying acceleration voltages (100, 400, 1200 kV) and an aperture of 3 mrad. [2]

$$\frac{d\sigma}{d\Omega dE} = \alpha^2 \cdot \frac{4 \cdot k_i^2}{\beta_i^2} \cdot \frac{k_f}{k_i} \cdot \frac{1}{2} \sum_{s_n, s_m} \left\{ \frac{1}{q^4} \left| < m | e^{i\vec{q}\vec{r}} | n > \right|^2 + \left(\frac{\vec{t}\vec{\beta}_i}{q^2 - \left(\frac{\Delta E}{\hbar c}\right)^2} \right)^2 \left| < m | \vec{\alpha}\vec{t} e^{i\vec{q}\vec{r}} | n > \right|^2 \right\}$$

Equation 1. Double differential cross-section with $\alpha \approx 1/137$ fine structure constant, $\boldsymbol{\beta}$ velocity of the incident particle normalised to the vacuum speed of light, **r** position of the atomic electron, $\boldsymbol{\alpha}$ vector of Dirac-matices, k_i wave vector of incident electron, q momentum transfer, **t** unit vector perpendicular to **q** and is situated in the plan of scattering process, $|n>$ initial state, $|m>$ final state and ΔE energy loss of the incident electron.