Transition probability functions for applications of inelastic core-loss scattering of fast electrons

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The quantum mechanical scattering of fast electrons on crystals has a long history. Especially in the transmission electron microscope (TEM), electron energy loss spectrometry (EELS) has a wide field of applications [1]. All of these methods have in common that the interpretation of measurements requires a thorough understanding of the theoretical framework and – most often – computer simulations.

For the core-losses discussed in this work, it is a common approach to write both the initial and the final states in the sample in spherical coordinates and subsequently perform a spherical harmonics series. As a result of this expansion, the weighted overlap

$$\langle j_\lambda \rangle(q) = \int_0^\infty u_i(r) j_\lambda(qr) u_f(r) r^2 dr$$

between the initial and final state wave functions of the sample enters the scattering equation. Here, q denotes the momentum transfer, j_λ Is the spherical Bessel function of first kind, and u_i, u_f are the radial wave functions of the initial and final state, respectively. Usually, this is expanded into a Taylor series for small q (the so-called dipole approximation), yielding

$$\langle j_\lambda \rangle(q) \propto q^\lambda.$$

Recently, it has been shown that this approximation fails even for moderate momentum transfers q [2]. One alternative to this is to calculate the wave functions with density functional theory (DFT), as is possible with, e.g., WIEN2k [3]. While this approach gives very accurate results, it is difficult to implement in numerical simulations due to the need to repeatedly perform numeric integrations of data points sampled on a (logarithmic) grid. In addition, the data is not accessible in an analytic form as is desirable for a better understanding of the physics involved.

In this work, we discuss improvements to the dipole approximation by using Slater-type orbitals (STOs) [4], as well as hydrogen-like orbitals (HLOs). With them, the weighted radial wave function overlap can be calculated fully analytically for any transition order λ [5]. The resulting formulas are only marginally more complex than the simple dipole approximation as they only involve polynomials (for HLOs) or polynomials and simple trigonometric functions (for STOs). Thus they are easy to implement in any existing simulation software currently based on the dipole approximation. Moreover, STOs and HLOs correctly model the asymptotic approach to zero of $\langle j_\lambda \rangle$ for large q, thereby ensuring particle number conservation.

The immense improvement over the first-order Taylor approximation is shown in fig. 1. It is clearly evident that both the STO and HLO calculations yield much better results than the dipole-approximation for dipole-allowed λ=1 transitions. For monopole transitions with λ=0, STOs fail because the nodes of the wave function close to the nucleus are neglected for STOs. HLOs, on the other hand, give a quite good approximation in that case as well.

References

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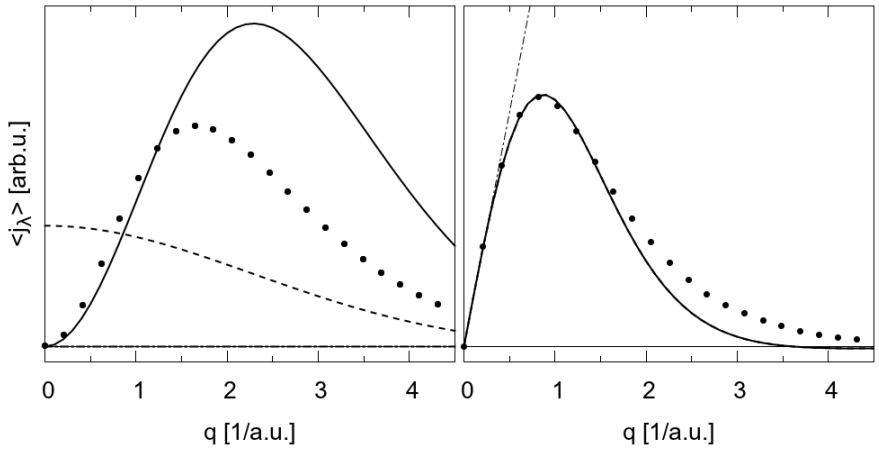

Figure 1. *Weighted radial wave function overlap $<j_\lambda>$ for monopole transitions (λ=0, left) and dipole transitions (λ=1, right) calculated with WIEN2k (dots), STOs (dashed lines), HLOs (solid lines). The dash-dotted line indicates the first order Taylor approximation as given in the text. Note that for these λ=1 transitions, the STO and HLO calculations coincide. Screening was taken into account by fitting to the WIEN2k data between q = 0 and q = 1.6 /a.u.*