Calculation of the angular-dependent loss function of Ag and Pd using ab-initio methods

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Electron energy-loss spectroscopy in the lower energy range (0-100 eV) yields information comparable to that of optical measurements but in contrast to optical spectroscopy is not limited to momentum transfer q=0. Theoretical calculations are of pivotal importance to help understanding the measured features. Density functional theory is a well established ab-initio method for the calculation of the electronic structure of solids and serves as a starting point for calculations of response functions.

In this paper we present theoretical studies of the angular-dependent loss function of silver and palladium together with experimental results. Our calculations have been performed with the developers' version of the "exc¡t!ng" code [1], which employs the LAPW basis to solve Kohn-Sham equations. Dielectric response was calculated in the independent particle approximation taking into account the induced Hartree potential (the so-called random-phase approximation, or RPA), which is sufficiently accurate for many metals. Local field effects were accounted for by considering at least three shells of reciprocal lattice vectors for the calculation of the macroscopic dielectric function.

The results for silver are presented fig. 1. For q=0 seven pronounced peaks in the 0-55 eV range are clearly seen. The peak at 3.2 eV (experimental value 3.8 eV) corresponds to the "classical" plasmon in Ag. The origin of other peaks has been identified as being caused by inter-band transitions from occupied *d* states to flat unoccupied bands which yield van-Hove-type singularities in the unoccupied density of states. The calculated positions and intensities of those peaks are in a very good agreement with most recent experimental results and substantially improve upon RPA calculations without local-field effects [2], especially at larger energies. We have analyzed the dispersion and damping of the "classical" plasmon [3], as well as the origin of the strikingly different dispersion of those peaks which originate from inter-band transitions.

The results for palladium are presented fig. 2, together with experimental data acquired on a monochromated tecnai [4]. The improvement gained by the inclusion of local field effects reduces peaks at high energies bringing the calculated spectra to a very good agreement with the measured ones.

References

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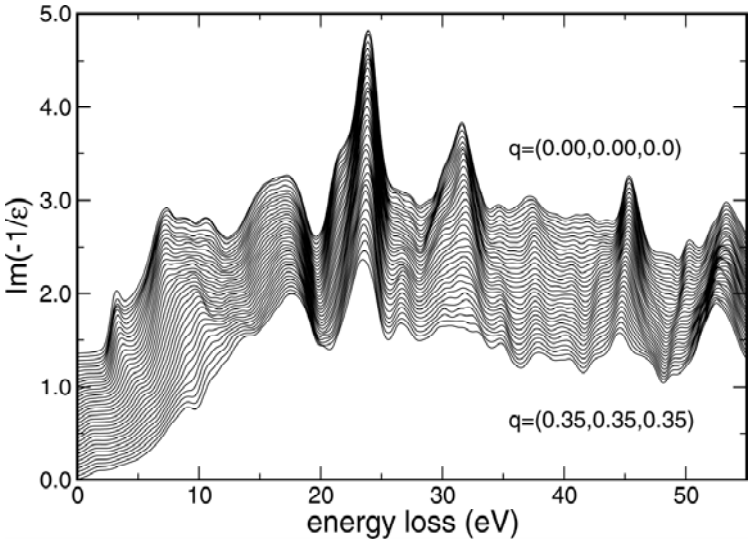

Figure 1. Energy-loss function of bulk fcc silver for momentum transfer q along the [111] axis. The highest curve corresponds to *q*=(0.0,0.0,0.0) a.u., while the lowest one corresponds to *q*=(0.35,0.35, 0.35) a.u. Spectra are offset for clarity.

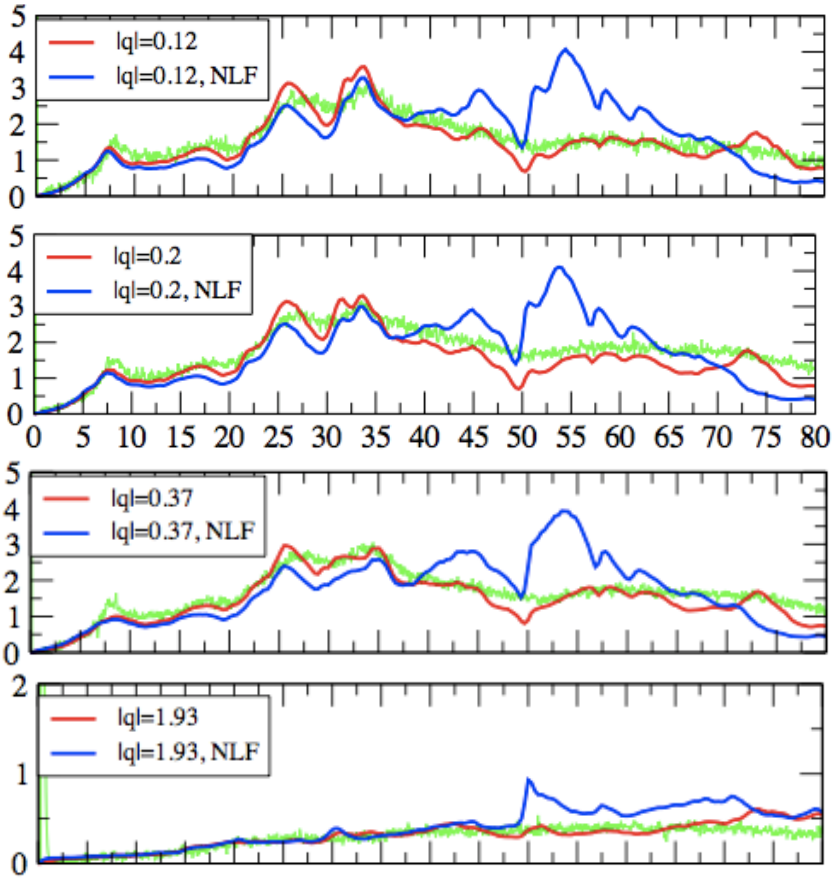

Figure 2. Energy-loss function of bulk fcc palladium for momentum transfer *q* along the [111] axis without (blue) and with (red, labeled NLF) local fields compared to experimental spectra acquired on a monochromated TEM (green) [3].

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