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Report:

We have performed high-resolution room temperature IXS measurements of the dynamical structure factor S($\mathbf{q},\omega$), proportional to $Im\ \varepsilon^{-1}(\mathbf{q},\omega)$, of silicon at several momentum transfers along the [100] and [111] directions. The aim of the experiment was to ascertain the importance of local-field and excitonic effects in the plasmon spectrum and to distinguish them from the features due to interband transitions, the latter being hardly resolved in previous IXS studies [1]. The measurements were motivated by preliminary first-principle calculations of the loss function using a new formalism based on a combination of many-body perturbation theory with time-dependent density functional theory on the one side, and a formalism using the local approximation for the exchange and correlation kernel (TDLDA) on the other side [2].

Measurements of S($\mathbf{q},\omega$) with x-ray photons of 7.9 keV energy and 200 meV energy resolution were performed for several q values along the [100] direction. The reduced number of sharp features found and the need of better statistics made us change to an energy resolution of approx. 1 eV, which was employed for most of the measurements.

Figure 1 shows a comparison between the measured spectrum for q=0.53 a.u. along the [100] direction (200 meV resolution) and that reported in Ref. [1]. The presence of a shoulder at 15 eV and a peak at 7 eV becomes evident in the new experimental data set. The shoulder can be attributed to exchange and correlation effects which are accounted for by our TDLDA calculation; only the

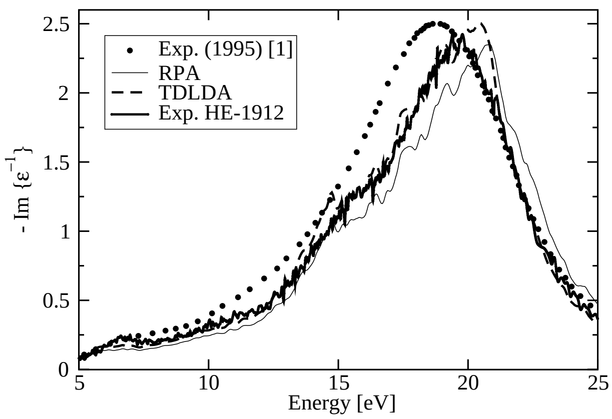

Fig. 1

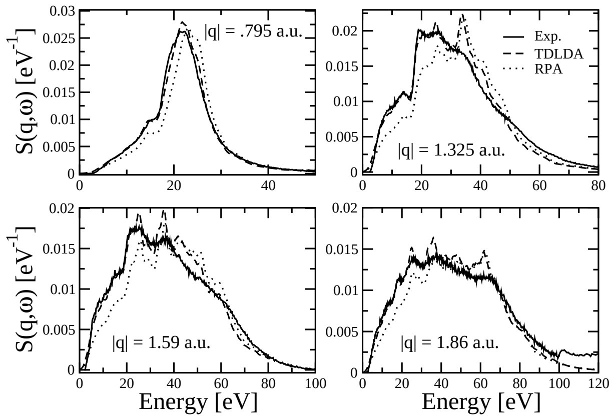

Fig. 2

interpretation of the small underestimate on the low-energy side of the main plasmon peak requires a more detailed calculation of excitonic effects which we are currently performing using the Bethe-Salpeter-derived TDDFT formalism. The simple RPA calculation yields relatively poor results.

Figure 2 summarizes $\mathrm{S}(\mathbf{q},\omega)$ measurements along the [111] direction. Since this is the bonding direction in silicon, a much richer fine structure is found for the plasmons. This has been already reported by Sturm and coworkers in Ref. [3], where they modeled the spectra with a Fano resonance resulting from the coupling of discrete plasmon states with the continuum of electron-hole-pair excitations. This resonance appears between 18 and 20 eV for q-values larger than 0.53 a.u. The analysis of the rich fine structure observed in these spectra, obtained with higher resolution than that used in [3], is still in progress. This figure also shows the excellent agreement of the measurements, q in [111] direction, with the TDDFT-LDA calculations. (A refinement of the analysis is still pending – fitting of the high-energy tail for the normalization along physical arguments; energy-dependent life-time broadening.) The agreement is consistently better than for the simple RPA calculations also shown.

Finally, we have performed tentative measurements of off-diagonal elements of the dielectric matrix using coherent inelastic x-ray scattering (CIXS), which has not been done since the pioneering work by W. Schülke and A. Kaprolat [4]. We have performed preliminary measurements for $\mathbf{G}_1 = (111)$ and $\mathbf{G}_2 = (220)$ and $\mathbf{q}_1 = 0.5(1,1,1) + 0.14(1,-1,0)$ and $\mathbf{q}_2 = (1,1,0) + 0.445(1,-1,0)$, following the recipe given in Ref. [4]. These preliminary measurements prepare future measurements of the off-diagonal elements, which will yield a unique opportunity to compare different theoretical approaches to calculate the dielectric response matrix.

References

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