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Advances in Momentum-Resolved Dispersion Investigations via Monochromated Electron Energy-Loss Spectroscopy

Michael R. S. Huang, Wilfried Sigle, and Peter A. van Aken

Max Planck Institute for Solid State Research, Stuttgart, Germany

While swift electrons pass through a thin film, various solid-state excitations occur through mutual Coulomb interactions, which convey a variety of useful messages relevant to the fundamental materials properties. Measurements by momentum-dependent electron energy-loss spectroscopy (EELS) directly access the dispersion relation, allowing the intrinsic band structure as well as the indirect (dipole-forbidden) transitions to be probed.

In this study, ω - q maps were acquired in the Zeiss sub-electron-volt-sub-angstrom microscope (SESAM), which is equipped with a monochromator and the advanced in-column Mandoline energy filter. Taking full advantage of this facility, which was dedicatedly designed for high dispersion and transmissivity together with superior isochromaticity, the revelation of further details of the characteristic dispersion is expected. Here an isotropic semiconductor Si (110) was used as a test example. During the experiment, the specimen was gradually raised above the eucentric height. In combination with varied magnifications in image mode, the effective camera length (ECL) can be significantly extended beyond the original specification limit, resulting in very high angular resolution [1].

Figure 1 shows the systematic variation of the ω - q map with various ECLs. The most prominent lines perpendicular to the energy-loss axis correspond to the excitation of the volume plasmon. With increasing ECL, the dispersions of electronic modes occurring at rather small scattering angles, for example surface plasmons (SPs) and retardation effects, become clearly resolved [2]. Another optimized ω - q map of Si, which reveals more detailed features, is given in Figure 2(a). In comparison with a result from the literature (Figure 2(b)) [2], all the main features reported are well reproduced in this experiment but with much enhanced energy and angular resolutions. The dispersion well below the bandgap onset (~3.4 eV) is related to the coupling of the surface guided modes to the bulk Čerenkov radiation [3]. For clarity, the theoretical calculation with a modeled Si slab of similar thickness to the experiment (approximately 100 nm) was performed by means of the relativistic Kröger equation [4]. As illustrated in Figure 3, quantitative consistency between the experimental data (colored circles) and the predicted profiles of scattering probability (black solid lines) can be reached, except for the SP. This deviation can be associated with the unavoidable formation of surface oxidation, which shifts the dispersion of SP toward lower energy. With this recorded map, the dependence of spectral profiles on momentum can be subsequently extracted by integrating the intensity over a given momentum width for further investigations.

In conclusion, the momentum and energy dependence of the low-loss spectrum of Si has been reexamined in the Zeiss SESAM. Distinct identification of the dispersive character with enhanced visibility in both large and small angular regimes confirms the suitability of this apparatus for more advanced characterizations. Based upon the above progress and improvement, the research direction in the near future will be orientated to more complicated anisotropic crystals, in particular two-dimensional materials, such as graphite/graphene or transition metal dichalcogenides, with emphasis on the exploration of their fundamental electronic and optical natures [5].

References:

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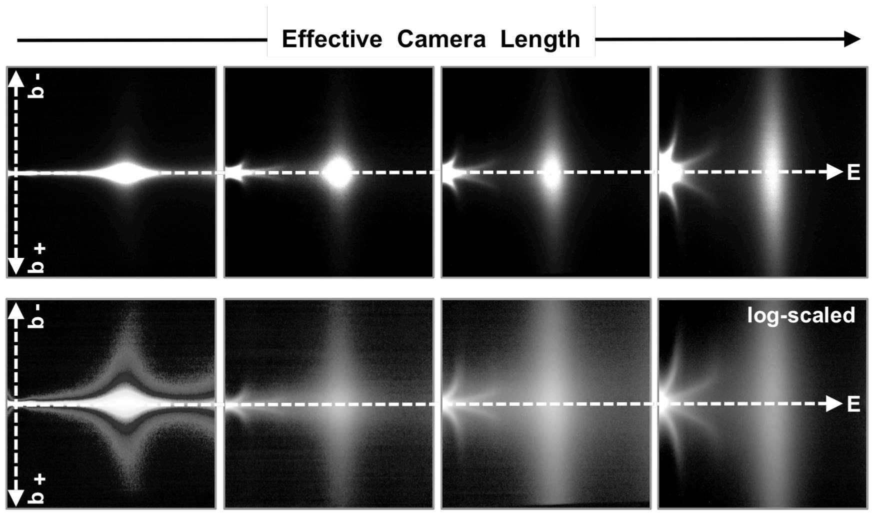
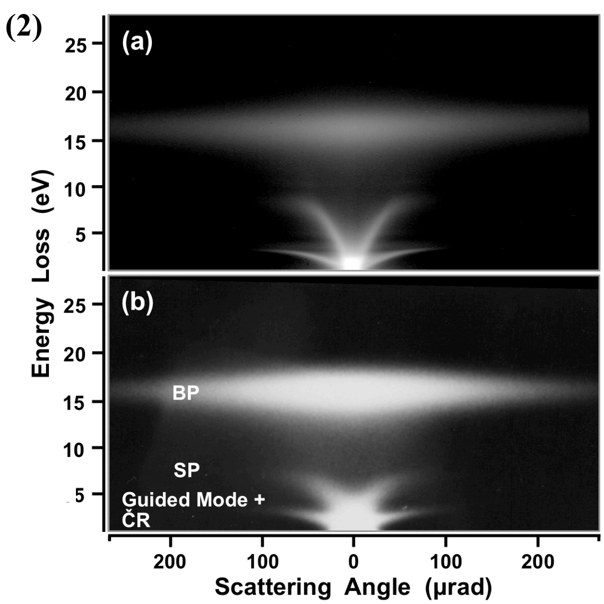


Figure 1. Systematic variation of the ω - q maps of single-crystalline Si with various ECLs.



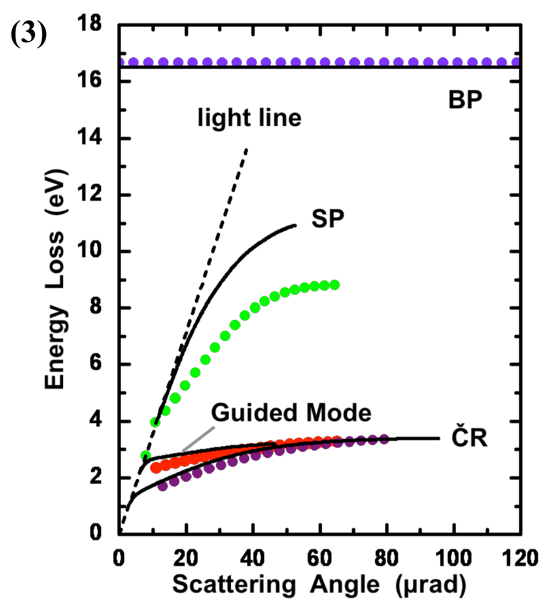


Figure 2. (a) The optimized ω - q map of Si compared to (b) a reference from the literature [2].
Figure 3. Comparison of the experimental data (colored dots) and theoretical profiles (solid black lines) calculated with the Kröger formula including relativistic contributions.