Spectroscopy in STEM/TEM

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Accessing low-energy cavity modes in rectangular slits in a thin Au film by STEM-EELS in combination with the Richardson-Lucy algorithm

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Scanning transmission electron microscopy in combination with electron energy loss spectroscopy (STEM-EELS) is becoming more and more important in the characterization of nanostructures due to its sub-nanometer spatial resolution which renders it far superior compared to optical methods. Even state-of-the-art optical methods like apertureless scanning near-field microscopy (SNOM) are limited to a resolution of 10 nm. Owing to sophisticated energy loss spectrometers or imaging energy filters in combination with electron monochromators and optimized acquisition techniques, an energy resolution better than 50 meV [1] is achievable. However, due to the tail of the zero-loss peak (ZLP), the accessible energy range is limited towards low energy losses which makes STEM-EELS disadvantageous compared to optical methods. The Richardson-Lucy (RL) algorithm [2, 3] was shown to improve the energy resolution of recorded spectra [4] by deconvolving the point-spread function of the detector system. In combination with electron monochromators, the RL algorithm offers the possibility to further improve the energy resolution and the accessible range of energy losses towards low-energy signals.

Recently, standing cavity-like modes hybridized with surface plasmon polaritions (SPP) were reported in rectangular slits in a thin Au film [5]. Additionally, strong enhancements of the cavity modes upon introduction of neighboring slits was demonstrated [6]. Due to this enhancement and the tunability of the cavity-mode energies by variation of the slit length, double slits are superb objects to evaluate the performance of STEM-EELS in the energy-loss range below 1.5 eV.

In this work slits were milled in a 200 nm-thin Au film using an FEI Strata 400S focused ion beam system. The experiments were performed in an FEI Titan³ 80-300 equipped with an electron monochromator (Wien-filter type) and a Gatan imaging energy filter (GIF) Tridiem model 865 HR. Under monochromatized conditions a probe size of about 5 Å is realized. The optimized acquisition procedure proposed by Bosman and Keast [7], binned gain averaging, was implemented to acquire high-quality EEL spectra. 50 spectra with exposure times of 1 ms were summed after energy drift correction and normalized with respect to the ZLP intensity. The RL algorithm was implemented with a self-written script in Digital Micrograph (Gatan). A reference spectrum recorded with the specimen retracted was applied as point-spread function for the deconvolution procedure.

Figure 1 shows spectra recorded within one slit with a size of 900 nm x 180 nm in an array of two slits separated by a metal bar with 100 nm width. A series of EEL spectra were acquired along the center of one slit (cf. arrow in the inset of Figure 1). The spectra are shifted vertically for better visibility. As the central bar separating the two slits is approached, two low-energy peaks are resolved at energy losses □E of 0.5 eV and 1.47 eV in addition to the surface plasmon peak at 2.3 eV. These are the fundamental mode and the 3rd harmonic of a cavity standing wave [5] along the long slit direction. Spectra taken close to the opposite slit wall (e.g. the black spectrum in Figure 1) also show these signals as very faint shoulders. The determination of the exact energy loss of these weak signals is difficult. Figure 2 shows how the application of the RL algorithm yields a significant improvement of energy resolution and visibility of these faint signals (for a spectrum recorded at 10 nm distance to the outer wall). With increasing number of iterations, the shoulder at □E = 0.5 eV is clearly resolved as a peak. The most significant effect of the RL algorithm is observed close to the ZLP which is strongly sharpened (inset in Figure 2). To check the effect of the RL algorithm, the same procedure was applied to a spectrum recorded at 10 nm distance to the central bar and compared to a spectrum which was only processed by subtraction of the reference spectrum. The results are depicted in Figure 3 which shows the raw spectrum, the reference spectrum, the spectrum obtained after 7 iterations with the RL algorithm, and the spectrum after reference subtraction. It is evident that the application of 7 iterations is, within the error limit, identical to the subtraction of a reference for □E≥ 0.5 eV. A power-law background fit to the spectrum after application of the RL algorithm can be used to evaluate the 0.5 eV peak.

Figure 4 shows spectra of three double slits with varying lengths, i.e. 900 nm, 1300 nm, and 2340 nm. Each spectrum was treated with 7 iterations prior to a bi-exponential background fit to the right side of the ZLP to recover the signals. As the slit length is increased, the energy of the fundamental cavity mode shifts to lower energies from 0.49 to 0.24 eV (cf. arrow). The small intensity to the left of the fundamental peaks is due to the background-fitting procedure. We also observe more excited high harmonics with increasing slit length. The shortest slit supports only excitation of the fundamental mode and 3rd harmonic, whereas even the 9th harmonic is excited for the longest slit. The ability to resolve a peak with an energy loss as low as 0.24 eV shows the viability of the RL algorithm for the evaluation of very low-energy signals with STEM-EELS.

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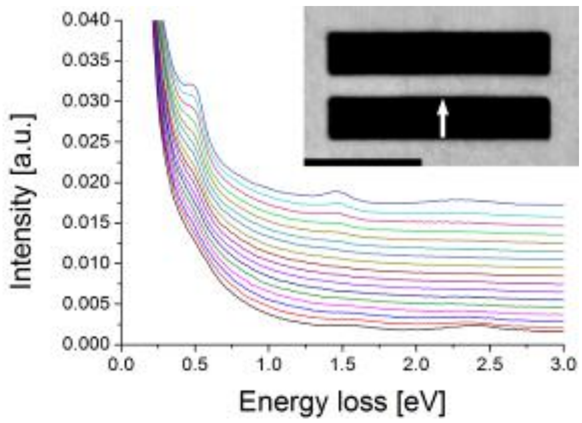

Figure 1. Series of EEL spectra acquired across one slit of double slit array. Inset: HAADF-STEM image. The arrow depicts the position (acquisition direction) of the spectra.

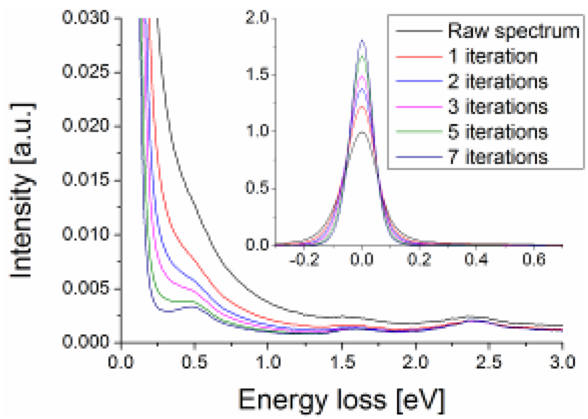

Figure 2. Effect of the RL algorithm on the bottom-most (black) spectrum in Figure 1. The faint signal at ΔE = 0.5 eV is clearly resolved with increasing number of iterations. Inset: Effect on the ZLP.

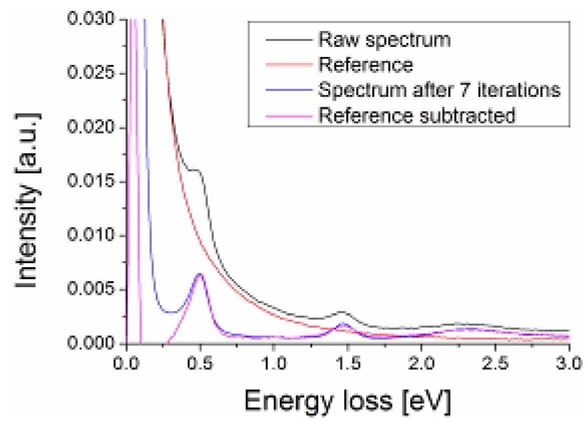

Figure 3. Comparison of the effect of the RL algorithm and reference subtraction to the top-most spectrum in Figure 1. Both methods show nearly identical results for ΔE exceeding 0.5 eV.

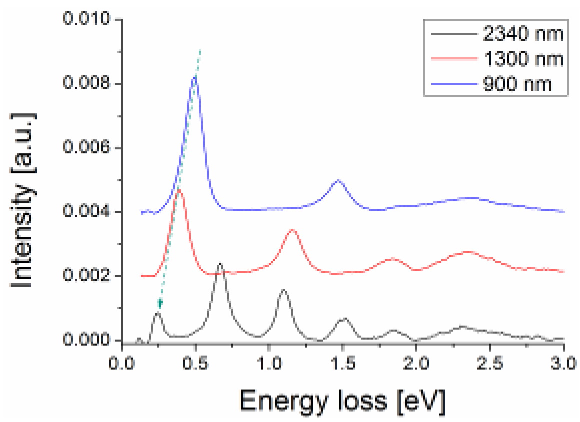

Figure 4. Spectra (after 7 iterations and a power-law background subtraction) acquired near the metal bar in double slit arrays of slits with varying slit lengths. Additionally to the red-shift of the fundamental energy, more high harmonics are excited with increasing slit length.