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In-situ Calibration for Angle-resolved Valence EELS

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Momentum-resolved electron energy loss spectroscopy (qEELS) in the optical and *x*-ray region, 1 to 10 eV, yields dispersion relations (ω-*q*) of the observed excitations. The ω-*q* relations assists identification of spectral features *q*EELS spectra [1,2]. Accurate calibration of energy dispersion in eV/pixel can be routinely obtained by several methods. However, reliable calibration of the momentum (*q)* dispersion at a camera length L = 10 to 100 m, that is used for valence *q*EELS, can be challenging. We report a method for *in-situ* calibration of *q* dispersion using the surface plasmon polariton (SPP) in a known material.

Fig. 1 shows an example of *q*EELS obtained from a ≈200 nm thick silicon sample [3]. A Hitachi HF-3300 TEM with a cold field emission gun and CEFID spectrometer [4] was utilized We collected 1,000 spectra over ~210 s interval at 300 kV. The spectra were aligned and summed using a Matlab™ script that was also for automated detection of spectral features and for fitting the SPP dispersion curve [5]:

$$\omega(q)^2 = \frac{\omega_P^2}{2} + c^2q^2 - \sqrt{\omega_P^4/4 + c^4q^4} \qquad (1)$$

Here ω_P is the plasmon frequency related to its energy $E = \hbar\omega_P$ where Planck constant $\hbar = 6.58\text{x}10^{-16}$ eVs, and *c* is the speed of light. In a material where SPP follows Eq. 1, only the knowledge energy calibration (horizontal axis in Fig. 1) is needed to obtain *q*-calibration in nm^{-1}/ pixel. In Si plasmon energy is ≈16.7eV, and $\omega_P = E_P/\hbar = 2.54\text{x}10^{16}$ Hz that can be used to verify spectrometer energy dispersion calibration, see Fig. 1. An example fitted SPP using Eq. 1 is in Fig. 2. The *light line*, i.e. the wavevector *k* of light propagating in free space $\omega_L = ck$, can be visually extracted from Fig. 1, to verify the *q*-calibration obtained from fit of Eq. 1. In the example given here, the *q*-calibration obtained from SPP fit (Eq.1) and from visually estimated light line (Fig. 1) agree within ≈6% one from another.

Fig. 2 indicates that the SPP can be detected over an energy range from ≈5 to 10eV. Below 5 eV, the presence Cerenkov radiation (CR) makes automated detection of SPP difficult while above 10 eV the SPP intensity, which decreases as q^{-3}, is too low to detect reliably. The surface plasmon at ≈16.7eV/√2 sets the upper limit where bound SPP can exist [5]. Regardless the limited energy range, typical *q*-dispersion values in *q*EELS yield a few tens of measured SPP locations, sufficient to perform a fit. The light line is on the inside of the SPP and CR, see Fig 1, marking the region where the SPP can not exist [2]. Although automatic detection of light line is difficult, it has the advantage of linear dependence on ω and at any given energy, only fundamental constants (ħ and *c*) are needed to extract corresponding *q* value and perform *q*-calibration. The SPP is affected by the presence of surface contamination but the light line that corresponds to propagation of light in vacuum is not affected by contamination when taken as the boundary with the smallest slope of ω–*q* relation.

The sample used for the *q*-calibration should be sufficiently thick to prevent coupling of the SPP on its surfaces that splits the SPP into symmetric and asymmetric modes. Care needs to be taken to ensure the film is perpendicular to the incident electron beam. A lack of symmetry between the +*q* and –*q* branches of the SPP would indicate that the sample surfaces are not perpendicular to the electron beam. A Si calibration sample with dimensions ≈200 nm (thickness) and few μm^2, fabricated by a focused ion beam instrument, can be easily placed on the same TEM grid as a studied sample, thus providing convenient calibration standard [6].

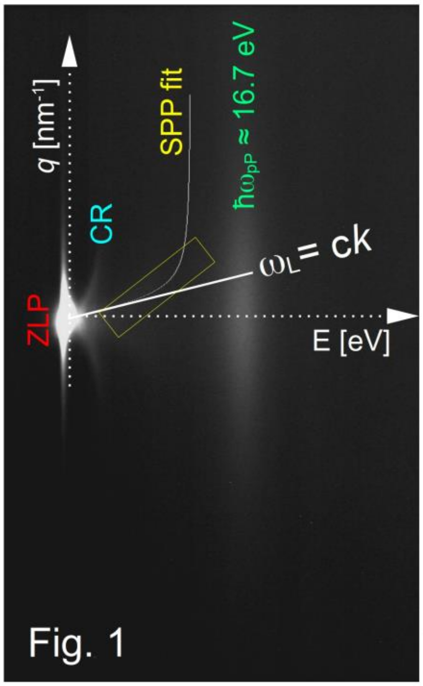

Fig. 1

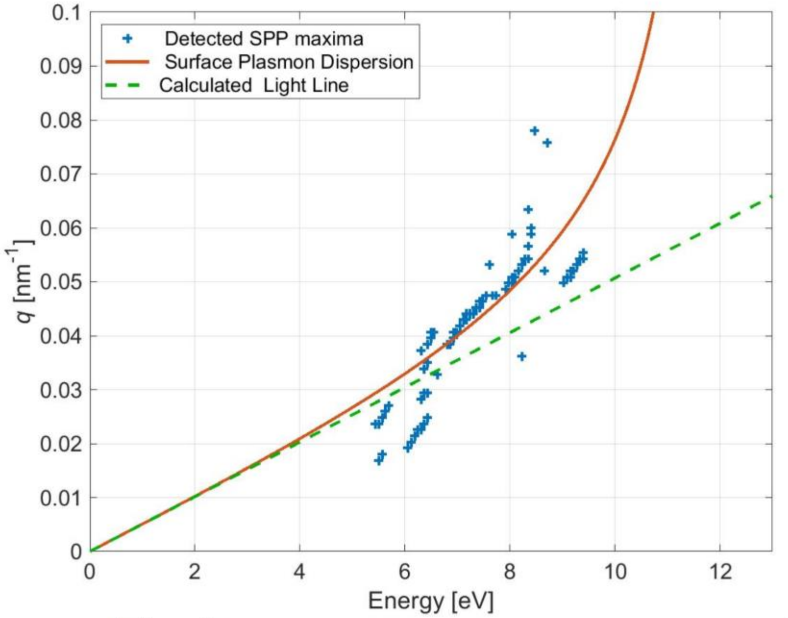

Fig. 2

Figure 1. An example of *q*EELS spectra from a Si slab acquired using Hitachi HF-3300 TEM operated at 300 kV with CEFID spectrometer. Horizontal axis is energy loss, vertical direction is momentum transfer. The marked spectral features are zero-loss peak (ZLP), Čerenkov radiation (CR), surface plasmon-polariton (SPP) fitted using Eq. 1, bulk plasmon at ω_P ≈16.7 eV. The light line $\omega = cq$ is marked by a solid white line. The full height of the spectrum corresponds to 1.16 nm^{-1} (–0.58 to +0.58) nm^{-1} corresponding to a maximum scattering angle ±1.1 mrad. The yellow rectangle marks the approximate area of automated search for SPP location.

Figure 2. Extracted location of the SPP (blue +), SPP fit (solid red line) and calculated light line $\omega_L = ck$ (dashed green line). The SPP was extracted by finding the maximum of the SPP spectral feature within the yellow rectangle in Fig. 1.

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