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STEM EELS Resolution Revisited.

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The ultimate resolution of scanning transmission electron microscope (STEM) images based on inelastic scattering is determined not only by the imaging optics, but also the delocalization of the inelastic scattering potential. This is particularly true of electron energy loss spectroscopy (EELS) where the scattering potential can be both quite broad and have long ranged tails. STEM EELS image resolution hence becomes related to the EEL spectra of the specimen constituents and is not simple a function of electron optics.

Two conceptually different measures are often used to describe the delocalization of images formed using the measurement of inelastic scattering. The first is the full width at half maximum (FWHM) of the image. This is commonly used for EELS images, since the scattering potential itself can be effectively nonlocal, and is hence not easily visualized [1, 2]. An alternative measure of delocalization is d_{50} which designates the diameter containing 50 % of the image intensity. These two measures are fundamentally different and should not be confused. For example in Fig. 1 we compare the measured d_{50} values of Lorentzian and Gaussian distributions with the same 0.75 Å FWHM. While the Gaussian has very similar values of the FWHM and d_{50}, the d_{50} of the Lorentzian is several times larger. Both these measures become somewhat more difficult to interpret if overlapping signals occur. The ability to resolve two overlapping signals is commonly defined by the Raleigh criterion R_{75} [3]. Figure 1 (b) and (c) show that the value of R_{75} is similar for both distributions.

Inner-shell ionization of course presents a far more complicated picture than these simple analytic distributions. The inelastic scattering potential is closely related to the reciprocal space transition matrix element describing the transition from the ground state 0 up to some excited state n

$$H_{n0}(\mathbf{q}) \propto \frac{\left\langle \psi_n(\mathbf{r}) \middle| e^{2\pi i \mathbf{q}\cdot\mathbf{r}} \middle| \psi_0(\mathbf{r}) \right\rangle}{|\mathbf{q}|^2}. \tag{1}$$

This term contains the details of both the initial and final state wave functions, which in turn define the energy loss. The momentum transfer $\mathbf{q}$ is depends on both the incident electron energy and the energy loss and its range is restricted by the detector geometry. Using this quantity we may calculate the scattering potential and subsequent "images" of isolated atoms. The FWHM, d_{50} and R_{75} can then be measured and compared. A simpler method of determining the d_{50} value for energy loss spectroscopy is based on the formulation of Egerton [4].

$$(d_{50})^2 = \left(0.61\frac{\lambda}{\alpha}\right)^2 + \left[0.5\lambda\left(\frac{2E_0}{E_{\text{loss}}}\right)^{3/4}\right]^2 + \left(0.6\frac{\lambda}{\beta}\right)^2. \tag{2}$$

Here the first term describes the probe diameter, the second term the dependence on incident energy an energy loss, and the third term includes the detector size.
A comparison of these delocalization measures is illustrated for O K-shell ionization and 100 kV incident electrons in Fig. 2. These are shown for different probe forming aperture semi-angles α as a

function of detector collection angle β. Provided $\beta > \alpha$ and the potential is essentially local, there is good agreement between the d_{50} values determined directly from calculated images and that calculated using Eq. (2). There are however significant differences between d_{50} and the FWHM in many cases.

In this presentation we will examine the variation of STEM EELS image delocalization for a range of incident energies and energy losses. We will also show the importance of the full quantum mechanical description, encapsulated by Eq. (1), when estimating image delocalization, most especially for energy losses near or below 100 eV where transitions from different core states show delocalization variations of up to a factor of four despite having similar threshold energies.

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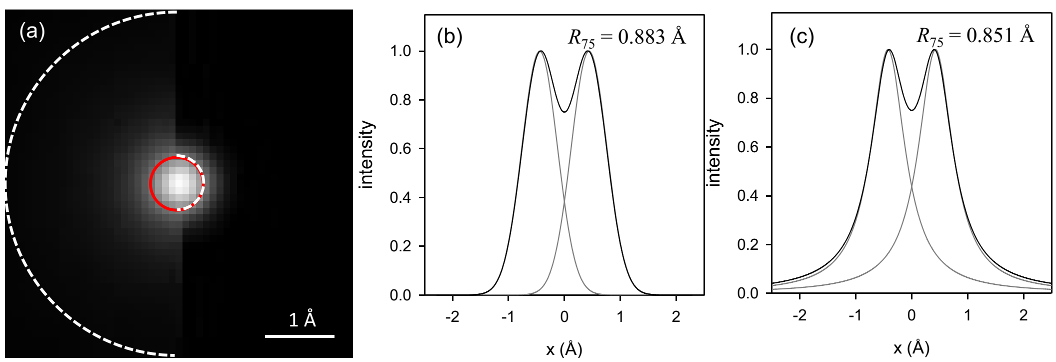

Figure 1. (a) Two dimensional plot of a Lorentzian (left) and a Gaussian (right). The FWHM (0.75 Å in both cases) is shown by the red circle and the measured d_{50} the white dashed semicircles. The Raleigh criteria resolution is shown by the black lines for a Gaussian in (b) and a Lorentzian in (c).

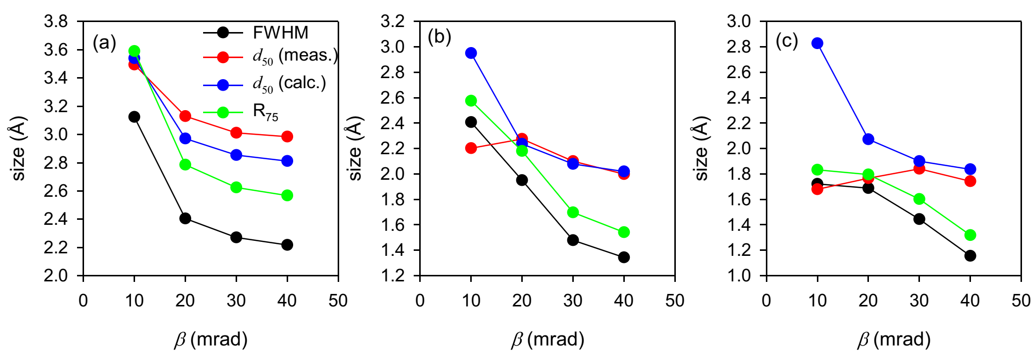

Figure 2. Comparison of the delocalization measurements for O K-shell ionization by 100 keV incident electrons. (a) α = 10 mrad, (b) α = 20 mrad and (c) α = 30 mrad. The FWHM is shown by the black lines. The value of d_{50} is shown by the red and blue lines for the measured and calculated values respectively. The green line shows the Raleigh resolution.

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