Inelastic STEM Imaging Based on Low-Loss Spectroscopy

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Aberration correction has led to improved resolution and sensitivity at lower accelerating voltages in the scanning transmission electron microscope (STEM). Imaging of beam sensitive two dimensional materials at atomic resolution has been enabled by operating at energies below the knock on threshold. In this way, single atom impurities have been imaged in BN using annular dark field (ADF) imaging [1] and electron energy-loss spectra (EELS) obtained in graphene with high spatial resolution [2]. The improved sensitivity has even made possible the measurement of energy-loss near-edge spectra (ELNES) providing information about local bonding of single impurity atoms [3]. We extend recent theoretical developments allowing the simulation of core-shell ELNES as a function of probe position to examine inelastic image formation based on low-loss spectroscopy [4].

In Fig. 1 we show an ADF image and simultaneously acquired low loss spectrum of graphene obtained using ORNL's Nion UltraSTEM100 operating at 60 kV. Spectrum images derived from the energy ranges of 20-26, 34-40 and 50-56 eV are also shown. The 34-40 eV image shows resolved atomic columns while the other images show no apparent contrast. It should be noted that the intensity variation is of the order of 5% and the image would be considered delocalized by most commonly used definitions. Such low levels of contrast are only visible due to the increased sensitivity of modern instruments.

The transition matrix element describing excitation from the valence band to the conduction band $\langle \phi_f | e^{2\pi i \mathbf{q} \cdot \mathbf{r}} | \phi_i \rangle$ can be used to construct the local inelastic scattering potential

$$V(\mathbf{r}) \propto \mathbb{F}^{-1} \left| \left\langle \phi_f \left| e^{2\pi i \mathbf{q} \cdot \mathbf{r}} \right| \phi_i \right\rangle \right|^2 / q^4 \quad , \tag{1}$$

where $\mathbb{F}^{-1}$ denotes an inverse Fourier transform. The image is calculated by convolving the probe intensity with the scattering potential. Correct modeling of the vacuum either side of the graphene sheets requires a large unit cell and a correspondingly large number of transition matrix elements.

Preliminary image simulations are shown in Fig. 2. Image simulations are shown with 5% noise added to show the effects of experimental noise on such low contrast images. While both of the lower energy images show graphene-like contrast, the addition of noise significantly reduces the visibility of the graphene structure at the lowest energy, while the graphene ring structure is still evident at the higher energy. The highest energy image does not show graphene like contrast. We will discuss the mechanisms underlying image contrast for inelastic STEM imaging based on low-loss spectroscopy.

References
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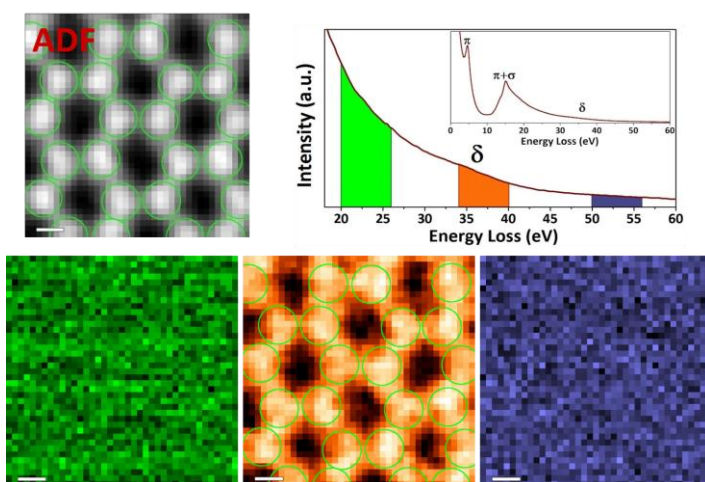[4] M. P. Prange et al., Phys. Rev. Lett. **109,** (2012), 246101.

Figure 1. Experimental ADF image and low-loss spectrum of graphene. Images derived from the indicated energy-loss regions of the spectrum are also shown. Scale bars are 1 Å.
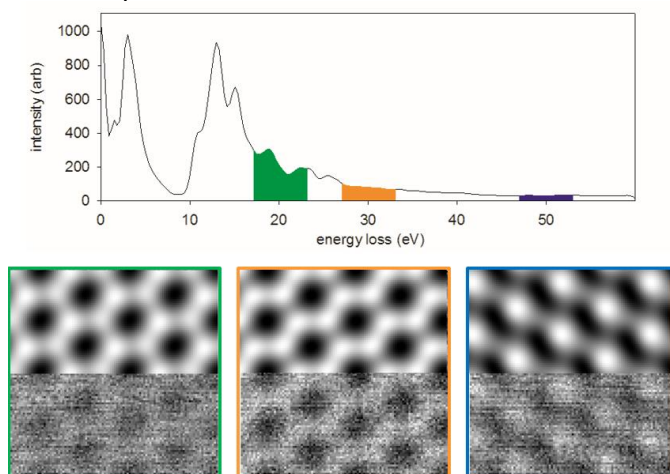

Figure 2. Theoretical spectrum and images derived from the indicated portions of the spectrum. Noise has been added to allow visual comparison with experiment.