

Prospects for Spatially-Resolved EELS of Atomic Point Defects

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A detailed understanding of point defects in materials, including their atomic structure and the induced local changes in electronic structure, is critical to the operation many established device materials (e.g., semiconductors), as well as many recently-proposed materials for quantum computing, cryptography and sensing (e.g., solid-state single-photon emitters [1]). On the other hand, our ability to probe the atomic and electronic structures of individual point defects arguably remains one of the grand challenges in materials characterization. The weak signals associated with individual point defects makes their detection intrinsically challenging. Hence advances in our ability to map their electronic states with spatial and energy resolutions that enable unambiguous identification and study will be greatly valued.

Here we examine the prospects of using electron energy-loss spectroscopy (EELS) in the scanning transmission electron microscope (STEM) to map the position of individual point defects via their electronic transitions (typically ~ 1 eV). EELS (supplemented with STEM imaging) allows us to probe the transitions which are important in many applications. Recent advances in electron monochromator and spectrometer instrumentation have opened the door to STEM-EELS at better than 10 meV resolution [2]. In the ~ 1 eV portion of the spectrum, these advances lead to increased detection capabilities for the discrete transitions relevant to many point defects. There are two main challenges: (1) the weak signals from individual point defects (and the implied beam damage), and (2) the relatively poor spatial resolution. The latter is implied by the degree spatial delocalization v/ω , where v is the incident electron's speed and ω is the energy loss. The delocalization is ~ 10 nm for energy losses ~ 1 eV (for a beam energy ~ 50 keV).

Figure 1 shows the (real part of the complex) charge density associated with an optically-active transition at a $N_B V_N$ defect (a nitrogen vacancy paired with a nitrogen substitution at the boron site) in single-layer h -BN. This defect is believed to play a major role in the reported single-photon emission from this material. The charge density was computed using density functional theory. The transition is that of an electron from the defect state (gap state) to the lowest unoccupied state. The transitioning charge is highly localized, to within a few nearest-neighbors, of the defect site. Also note the strong in-plane dipole character of the charge density with respect to the defect site, indicative of an optically-allowed transition.

Figure 2 shows the corresponding Møller potential, i.e., the EELS scattering function [3]. The Møller potential displays a width of ~ 10 nm despite the charge density being easily localized to within 1 nm. Hence the predicted spatial resolution is ~ 10 nm. This spatial resolution is, in fact, an order of magnitude better than the wavelength-limited resolution available in optical spectroscopy, e.g., photoluminescence, and would thus offer a significant improvement for defect mapping. Moreover, such delocalization could, in fact, be advantageous in that it could allow for minimal momentum transfer to the transitioning electron, thus minimizing inadvertent changes in the defect's charge state.

The theory described here is applicable to point defects in a broad range of materials, and many of the statements above apply will apply to other optically-active point defects. The theory is currently being utilized to interpret EELS experiments on materials for quantum and energy applications [4].

References:

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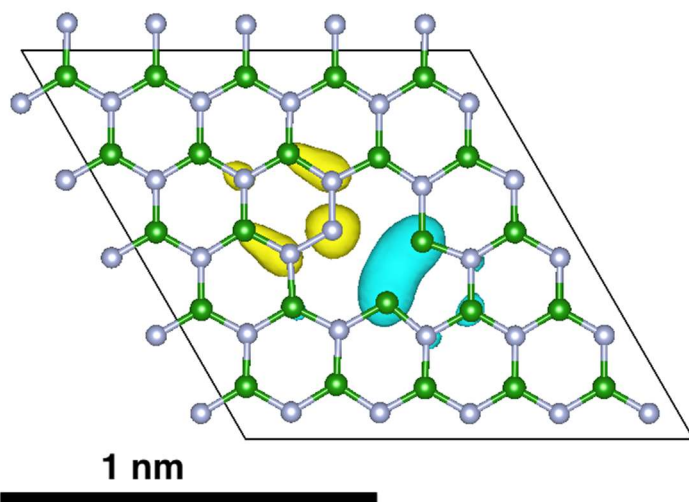


Figure 1. The (real part of the complex) charge density associated an optically-allowed transition between electronic states of a $N_B V_N$ defect in single-layer *h*-BN. The atomic structure is superimposed; nitrogen=silver, boron=green, positive charge=yellow, negative charge=cyan.

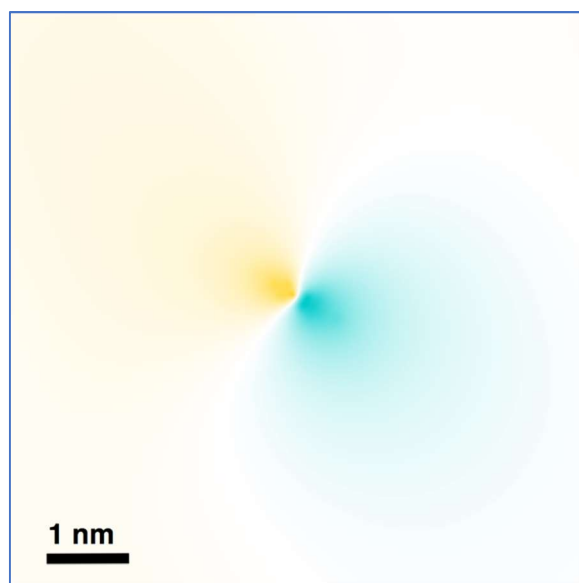


Figure 2. The Møller potential (EELS scattering function) for the transition in Figure 1, exhibiting inelastic delocalization of ~ 10 nm (defect at the center); positive=yellow, negative=cyan, zero=white.