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Identification and Manipulation of NV Centers in Nanodiamond

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The ability to unambiguously identify and directly position single-atom defects is important to the design and function of quantum technology devices. Color-centers in diamond are known single photon emitters (SPE) with applications in biomarking and quantum communication. Nanostructured diamonds, such as nanodiamonds and thin films, are the preferred morphologies for such applications. The properties of the nitrogen-vacancy (NV) center in nanodiamond, in which a N atom occupies a diamond lattice site with a neighboring lattice vacancy, is dependent on the defect charge state and nanodiamond surface, and directly probing a single defect is therefore important. Aberration-corrected scanning transmission electron microscopy (STEM) offers a sub-Angstrom diameter probe for single-atom sensitivity imaging and spectroscopy that can be used to investigate defects in nanostructured diamond.

Using a Nion UltraSTEM-X operated at 60 kV with simultaneous electron energy loss spectroscopy (EELS) and energy dispersive X-ray spectroscopy (EDS), we are able to identify single NV centers in 2-5 nm meteoritic nanodiamonds with EELS detection of the characteristic NV center peak at 282.4 eV [1] and the N signal in EDS [2,3]. Single-defect identification becomes more challenging when studying commercially-available, technologically-relevant synthetic nanodiamonds with diameters of 10-30 nm. As the sample thickness increases, the sensitivity to detect single-atom defects decreases, and it is likely that multiple NV centers need to be present within a single spectrum image pixel for the defects to be detected. Thus, while we do detect NV centers in the larger, synthetic nanodiamonds, isolated single NV centers may still be present and undetected. In addition to identifying single-atom defects, the directed positioning of single-atom defects in materials with application in quantum technologies is of interest. In recent years, STEM beams have been used to manipulate single atoms in 2D and 3D materials, advancing beyond the limited surface capabilities of scanning tunneling microscopes (STM) and atomic force microscopes (AFM). At 60 kV, the maximum energy transferred to N in a single elastic collision is 9.95 eV. This is above the migration energy of an NV center (4.9 eV) or interstitial N (1.8 eV) in diamond [4]. It should therefore be possible to use the focused STEM probe to directly position N defects in a nanodiamond. Shown in Figure 1, by scanning the beam over a single nanodiamond for an extended period of time, we demonstrate the ability to: 1. gather N atoms into one region of the nanodiamond; and 2. redistribute the N throughout the nanodiamond. Nitrogen EDS maps are shown in Fig. 1e-f, where the brighter pixels indicate higher N counts. In Fig. 1e, the N concentration has increased in the slow-scan direction, suggesting the prolonged scanning of the e-beam has swept the N atoms into one region. By rotating the nanodiamond 180º and scanning again for 3 hr at 24 μs/pix, the N atoms are redistributed into the nanodiamond (Fig. 1f). This is a significant step toward single-defect manipulation in nanodiamond [5].

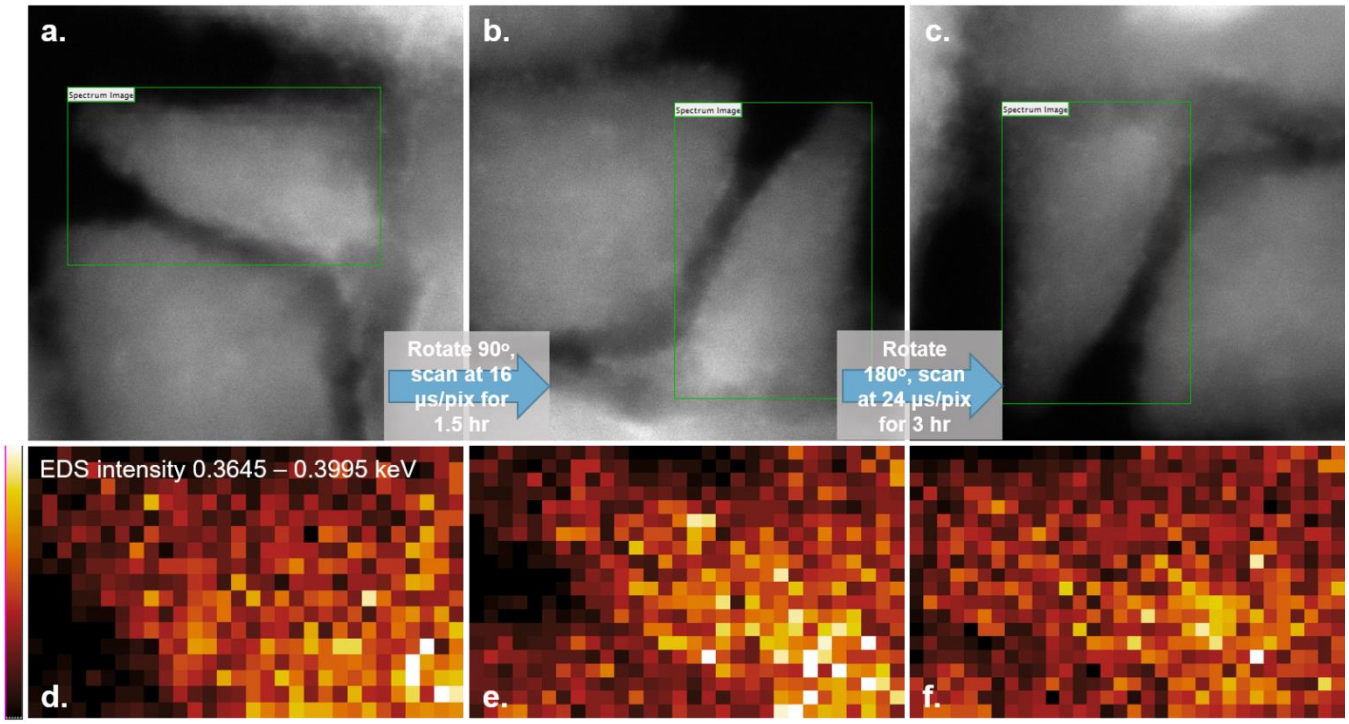

Figure 1. Using the STEM beam to move N atoms and NV centers in a nanodiamond. (a) HAADF image of nanodiamond prior to N movement. (b) Orientation of nanodiamond for 1.5 hours of scanning at 16 µs/px. (c) Orientation of nanodiamond for 3 hours of scanning at 24 µs/px. (d) EDS map centered on N intensity displaying N position prior to extended periods of scanning. (e) EDS map of N after 1.5 hours of scanning. N atoms (brighter pixels) have gathered in the lower right corner. This is in the slow-scan direction of the electron beam, indicating that N have been "herded" by the beam. (f) EDS map of N after rotating the nanodiamond 180º and scanning for 3 hours. The bright concentration of N is no longer present, and the N atoms have been distributed into the nanodiamond.

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