Transmission electron microscopy and spectroscopy for high spatial resolution color center detection and correlation

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Fluorescent Nanodiamonds (FNDs), diamond particles containing atomic defect color-centers, draw widespread attention due to their exceptional optical and spin properties, which make them excellent candidates for room-temperature quantum sensing and biomedical diagnostic applications. However, it is now recognized that continued advances of FND applications mandate a precise understanding of the crystallographic environment around the defect centres as well as the surface properties of the particles themselves. These factors have been speculated to strongly affect their fluorescent brightness and optical stability. Moreover as most of the FND fabrication is a top-down process where larger diamond crystals are milled to desired nanometer size, FNDs generally have broad size distribution and irregular shapes and have shown to exhibit as large as 7 fold brightness variations for same nominal size. Hence, an understanding of the electronic structure of FND surfaces, and ultimately how to control them, becomes important for FND applications.

Conventionally optical microscopy and a range of spectroscopies including fluorescence microscopy, Raman and electron paramagnetic resonance (EPR) are valuable in measuring different types of color centers in diamond. However due to the wavelength limitation, the optical measurement methods generally have resolutions of ~hundreds of nm. In order to achieve better understanding of FND properties, transmission electron microscopy offers better resolution and capabilities to provide insights at atomic resolution. My group has been developing methods using electron energy loss spectroscopy (EELS) in a TEM to detect and locate color center related defects, and the surfaces structures of FNDs. In particular, our results demonstrate that locations of NV centers can be measured at better than 1nm resolution. In addition the generally poorer signal-to-noise of EELS is ultimately compensated by the highly localized (subnanometer) probe, which enables unambiguous spectroscopic data obtained directly from the FND surfaces. By directly correlating with the fluorescence microscopy map, our newly developed correlative TEM and fluorescence microscopy (TEMPL) demonstrates that the 3D shapes of FND have strong effect to their brightness up to 4 folds enhancement for FNDs $< \sim 100$ nm^3. Such direct electron microscopy and spectroscopic data in being able to distinguish between competing effects within complex FND, is valuable in advancing FND production and applications.