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Preparation and Characterization of Eu-Doped Diamond Samples by Atom Probe Tomography

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Practical quantum technologies built from wide band gap semiconductors, in particular diamond and silicon carbide, have become realistic thanks to careful defect engineering of these materials. Color centers, created by point defect dopant atoms, such as silicon and nitrogen, are the basis for nanomagnetomers that can sense and measure the state of a single nuclear spin and provide a platform for the entanglement of macroscopically separated spin states. The advancement of diamond quantum technologies will require better analytical techniques to probe the distribution of dopant atoms in these materials. To achieve optical addressing of individual spins, the dopant levels in the materials must be quite low, consequently traditional nano-analytical techniques such as STEM-EELS and STEM-EDS cannot provide the required chemical sensitivity to understand dopant distributions in these materials.

Local electrode atom probe tomography (LEAP) provides 3D spatiochemical mapping with elemental sensitivity down to 1 ppm and sub nm spatial resolution. Recently, LEAP has proven invaluable for analyzing dopant concentrations in FIN-FETs, where the 3D doping profile is critical to the performance of the devices and there may be only a handful of dopant atoms per fin structure. Atom probe tomography of diamond remains challenging due to the low conductivity and large band gap of the material, however the insights into dopant clustering and local dopant environment could provide information regarding the selection of growth, doping, and processing parameters, consequently leading to better materials and hence better devices.

Recently the incorporation of europium defects in to diamond was reported [1]. Europium is notable because it can exhibit optically detectable nuclear magnetic resonance and is known for its particularly long nuclear spin coherence times. In diamond, Eu has the potential to act as a long term quantum memory to store spin information. Currently, the exact nature of the Eu in the diamond remains unknown. Atom probe tomography of this material can provide insights into whether the Eu exists as the requisite isolated defects, as clusters, or even as nanoparticulate inclusions. A High resolution STEM EDS mapping of the Eu-doped diamond was unable to chemically identify any Eu present in the material. Atom probe is the only technique that can provide tomographic elemental information on the length scales necessary to identify the nature of the europium in these diamond samples; information that is critical to gauging the potential utility of this material as a quantum memory and for other applications.

Single crystal CVD grown diamonds were prepared [1] and the classical lift-out method has been used to produce APT samples. It has been noticed that the use of a C-weld instead of a Pt-weld greatly improve the success of APT experiments. Diamond is known to induce the formation of 'pillars' under ion milling, and in the case of our samples, these diamond pillars have a size in the 20-50 nm range, sufficient for further APT analysis (**Figure 1a**). Experiments were performed in a LEAP 4000X HR system with conditions of acquisition: 60-80 pJ, 100 kHz, 50 K, DR: 0.2-0.5%. The presence of carbon clusters up to C_{20} is currently observed in the mass spectra. However, these specific conditions of sample preparation and acquisition allow collecting a few millions of atoms before fracture of the sample, and thus allow a relatively decent reconstruction of the diamond sample (**Figure 1b**). Different sample preparation techniques and possible way to improve the analysis are also discussed in this contribution. [2]

References:

[1] Magyar, A. et al. Synthesis of luminescent europium defects in diamond. *Nat. Commun.* 5:3523 doi: 10.1038/ncomms4523 (2014)
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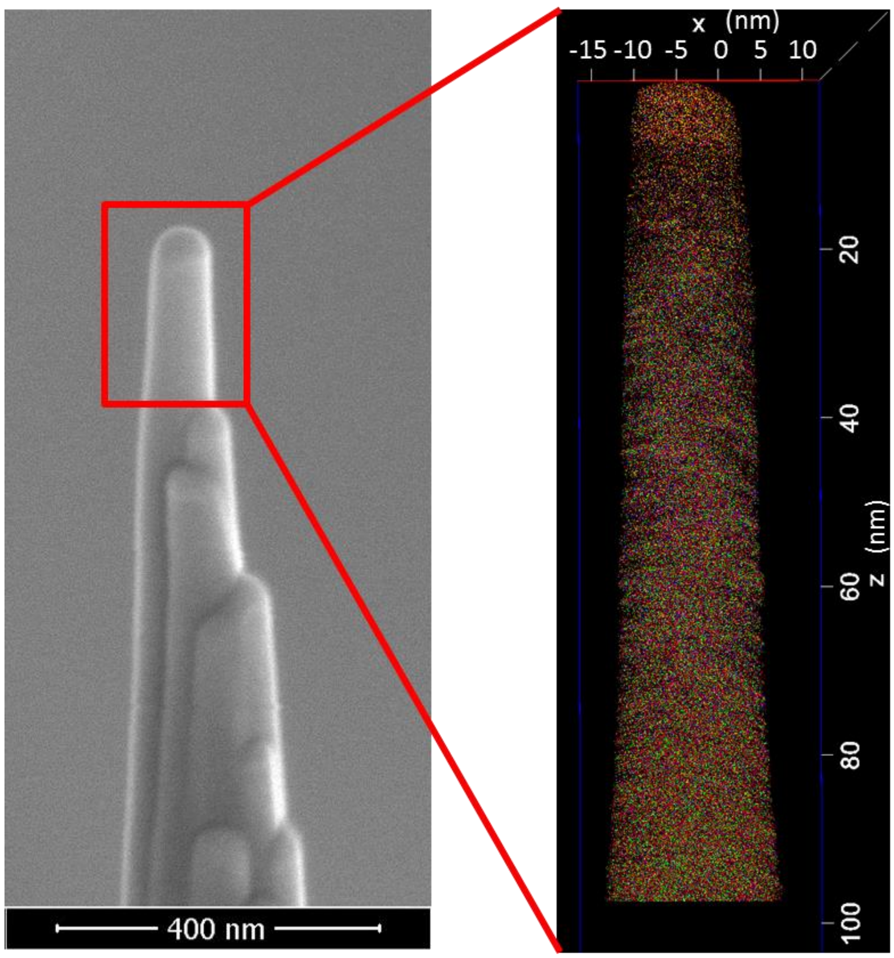

Figure 1. a) Typical single diamond 'nano-pillar' obtained after ion-milling and used for APT analysis b) 3D reconstruction of a diamond sample representing C clusters from 'C_1' to 'C_5'.