**Diamond Surface Functionalization via Visible light-Driven C–H Activation for Nanoscale Quantum Sensing**

Nathalie de Leon
Department of Electrical and Computer Engineering
Princeton University, Princeton, NJ, USA 08540

Nitrogen-vacancy (NV) centers in diamond are a promising platform for nanoscale nuclear magnetic resonance (NMR) sensing. Despite significant progress towards using NV centers to detect and localize nuclear spins down to the single spin level, NV-based spectroscopy of individual, intact, arbitrary target molecules remains elusive. NV molecular sensing requires that target molecules are immobilized within a few nanometers of highly coherent NV centers. The inert nature of diamond typically requires harsh functionalization techniques such as thermal annealing and plasmas, limiting the scope of functional groups that can be attached to the surface. Solution-phase chemical methods can be more readily generalized to install diverse functional groups, but they have not been widely explored for single-crystal diamond surfaces, and their compatibility with shallow NV centers has not been established. In this work, we report a versatile strategy to directly functionalize C–H bonds on single-crystal diamond surfaces under ambient conditions using visible light. Hydrogen atom abstraction of C–H bonds generates carbon-centered radicals that can intercept various radical acceptors to form C–F, C–Cl, C–S, and C–N bonds at the surface. We verify covalent bond formation using lab-based and synchrotron-based surface analysis. This functionalization method is compatible with coherent and charge stable NV centers within 10 nm of the surface with spin coherence times consistent with state-of-the-art, and we use shallow ensembles of NV centers to detect nuclear spins from functional groups attached to the surface. Our approach to surface functionalization based on visible light-driven C–H bond activation opens the door to deploying NV centers as a broad tool for chemical sensing and single molecule spectroscopy.