Engineering the Charge Occupancy of Nitrogen Vacancies in Diamond

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Introduction:

Nitrogen vacancy (NV) centers are point defects in a diamond lattice that occur when a carbon atom is substituted with a nitrogen atom while a neighboring carbon atom is removed entirely. The resulting gap in the lattice has many desirable properties, such as paramagnetism and optical energy level transitions, that make it an ideal candidate for single-photon optics and quantum computing applications [1]. But the NV center tends to lose one of its surrounding electrons due to recombination with wandering positive charges in the crystal, providing a damaging barrier to the development of large-scale, multipartite quantum networks.

This project sought to deter this process by depositing transparent metal oxides and films on the surface of diamond nanowires, with a goal of inducing charge discontinuities at the surface that would stabilize the electronic configuration of the lattice by inducing a negative charge excess in the region around the defect. Additionally, metal oxide semiconductor field effect transistor (MOSFET) devices were designed that used a voltage gate on the surface of bulk diamond to accomplish the same effect, albeit with more control of the dopant level in the region due to modulation of surface voltage [2]. Designs and fabrication processes for both devices were created and ensemble measurements of devices using confocal microscopy have begun.

Procedure:

Nanowire Fabrication. High-temperature, high-pressure Type Ib diamonds were obtained (HPHT, Element Six) and polished, and surface contaminants were removed with a boiling bath of equal parts perchloric, sulfuric, and nitric acid. 1%XR was spin-coated onto the diamond surface, followed by equal parts FOx-16 and methyl isobutyl ketone resists. Arrays of disks ranging from 200 nm to 250 nm in diameter were then patterned with varying dosages using an electron beam lithography system (Elionix ELS-F125). Tetra-methyl ammonium hydroxide was used to develop the resist, and reactive ion etching was used to carve away diamond from around the disk patterns, creating pillars roughly 1.5 microns in height. Excess resist was removed using boiling Piranha etch (80% sulfuric acid, 20% hydrogen peroxide). Surface oxides were then deposited using atomic later deposition (50 nm SiO_2 and 16 nm Al_2O_3), and the nanowires subsequently underwent rapid thermal annealing (three hours in O_2).

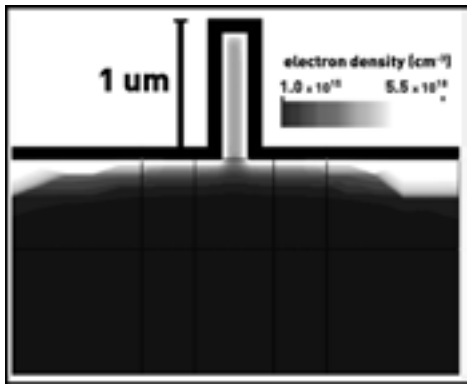

Figure 1: A finite element simulation of doping in a diamond nanowire due to a surface coating of silicon dioxide.

MOSFET Fabrication. Diamonds were obtained and cleaned in the same manner as the nanowire samples. Using plasma-enhanced chemical vapor deposition, the polished surface was coated with 400 nm of silicon dioxide, onto which HDMS primer and SH1813 photoresist were spin-coated consecutively. Exposure was performed using direct write photolithography on a Heidelberg uPG501 with typical doses in the range of 100mJ. The patterned sample was then developed and placed in buffered oxide etch for two minutes. After cleaning and optical inspection, the sample was coated with 300 nm lift-off resist LOR 3 3A followed by 500 nm SH1805. A second mask was then aligned to the visible oxide pattern and exposed with the same dosage. The SH1805 was developed, and the sample was then covered with 150 nm of gold using electron beam evaporation. The assembly was left overnight in Remover-PG at 80°C for gold lift-off. After the gold contacts were inspected and the sample was cleaned, 500 nm indium tin oxide was deposited on the sample using sputtering (AJA Orion 3) and then topped with HDMS/SH1813 photoresists in the same manner as the first layer. A final mask was aligned and exposed, and the ITO layer was then chemically etched using a mixture of equal parts concentrated hydrochloric acid and water.

Results and Future Work:

Based on numerical models, devices were designed and fabricated that locally doped the diamond surface by donating free electrons into the diamond conduction band. Nanowires [3] were chosen to receive passivation coatings because they had a high

ratio of surface area to volume, allowing maximal theoretical carrier concentration around the vacancy center (Figure 1).

After recipe optimization, wires were successfully created on the surface of bulk diamond that exhibited satisfactory deposition uniformity for two different oxides (Figure 2), suggesting that the method successfully can be adapted for a variety of different oxides and thicknesses.

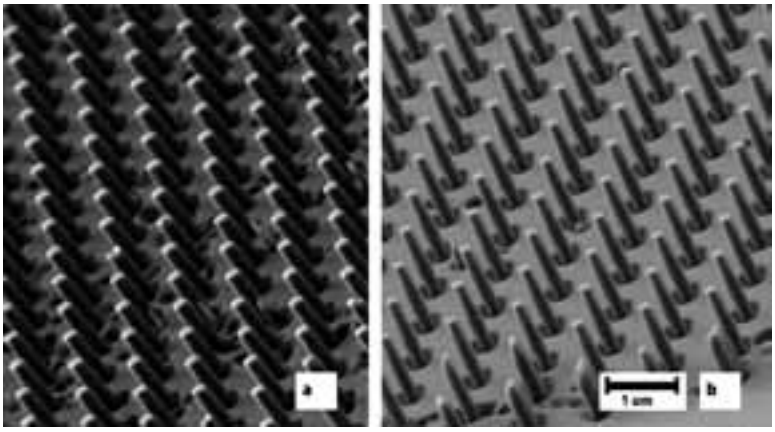

Figure 2: Diamond nanowires that have been coated with (a) 50 nm silicon dioxide, and (b) 22 nm aluminum oxide.

MOSFET devices were designed that adhered to material parameters determined by numerical simulations while allowing experimental flexibility. A three-mask photolithography process, including a gold lift-off step, was designed and optimized to yield high fidelity to the original digital design (Figure 3). While the process was originally developed on a silicon substrate for convenience, some difficulty was encountered in adapting the method for a smaller (2.5 mm square) diamond substrate, in part due to difficulty focusing and aligning on a smaller sample. These problems are purely technical in nature, and are expected to be resolved by the use of a specialized sample holder for the diamond during the lithography process.

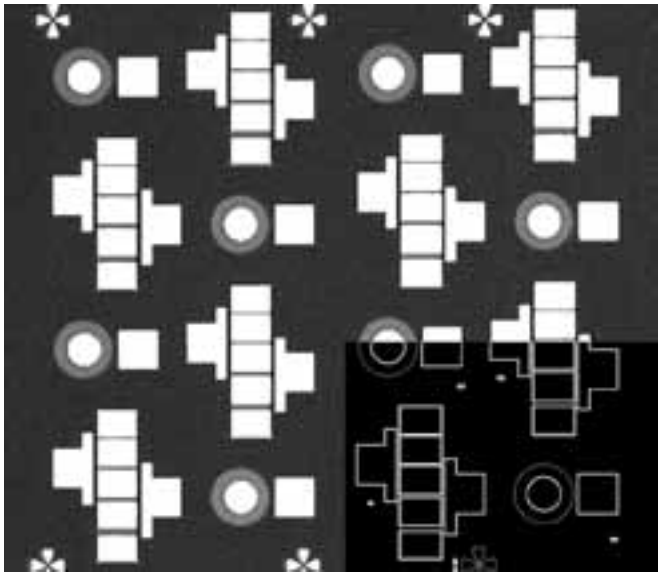

Figure 3: MOSFET structures on silicon. The smaller disks are 60 µm in diameter, and the original CAD design is inset.

Anti-correlation was successfully measured in photons emitted from the devices, and the nanowires yielded a clear fluorescence signal under laser excitation (Figure 4). Repeated measurements of ensembles of each type of device, however, are necessary to determine whether there is a significant increase in fluorescence from diamonds with devices, which would signal successful charge state stabilization of NV centers.

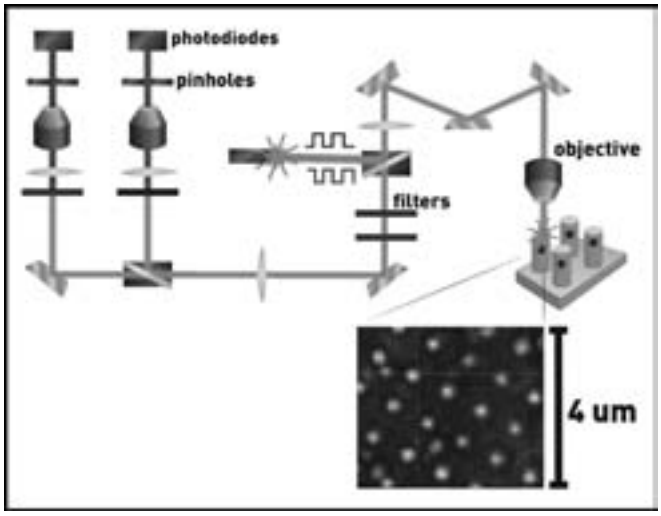

Figure 4: The confocal microscopy measurement setup, with a sample image of NV centers in nanowires.

Acknowledgments:

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References:

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[3] Babinec, T.M., et al.; "A diamond nanowire single-photon source"; Nature Nanotechnology, 5, 195-199 (2010).