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Abstract: We demonstrate a fabrication technique for high volume production of high-quality nanocrystals from bulk chemical vapor deposition diamond. Ramsey and Spin-Echo measurements confirm the long spin coherence of nitrogen vacancy centers in these nanocrystals.

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The spin properties of the negatively charged nitrogen vacancy center (NV) in diamond have made it the focus of many recent investigations across a broad range of fields [1]. Prominent among these are works in quantum-enabled sensing, where the NV is used as a local magnetic [2] and electric [3] field sensor. In addition, there is interest in using NV-containing nanocrystalline diamond as a biological labeling agent due to its cytocompatibility, fluorescent brightness and suitability for super-resolution imaging [4, 5] . Combining imaging studies with high-performance sensing requires high purity nanocrystals with good spin properties. However, commercially available detonation and high-pressure high-temperature diamond nanocrystals are of lower quality than the bulk chemical vapor deposition (CVD) diamond used in the sensing proof-of-concept experiments, substantially diminishing the phase coherence time of the NV center and therefore its suitability for quantum information and sensing applications [6]. Here, we demonstrate a method by which we produce a high yield of NV-containing CVD diamond nanocrystals, allowing for a substantial increase in NV phase coherence time and enabling the use of diamond nanocrystals as high-sensitivity field sensors and long-coherence spin qubits.

Fabricating nanocrystals directly from bulk diamond presents several challenges. Traditional electron beam lithography and oxygen reactive ion etching (RIE), which has been used extensively to create diamond nanostructures, stuggles to reach resolutions of lower than 50 nm with the smallest structures demonstrated in diamond having dimensions of 75 nm [7]. In addition, fabricating large quantities of such structures at high resolution across a macro-scale diamond substrate would be extremely time-consuming. We resolve these issues by using a self-aligned masking process, in which nanoscale metallic grains serve as an etch mask. The metallic grains can be extremely small and deposited with high density across a large surface in parallel, for example via sputtering. Ordinarily, the small size of these seeds would limit the aspect ratio and ultimate size of the resulting structures. However, due to the anisotropic nature of the plasma in the oxygen RIE that we perform, we achieve a final size and aspect ratio that is suitable to host NV centers for use as sensors.

After fabrication, we induced NV center formation through ion implantation and annealing, and mechanically separated the diamond nanocrystals from the bulk. We then proceeded with optical measurements using confocal microscopy and pulsed optically detected magnetic resonance (ODMR) to determine the spin properties of the CVD nanodiamonds. After identifying single NV centers through confocal scans, fluorescent spectra and second order autocorrelation, we measured the characteristic coherence times of the system, T_2^* and T_2, via Ramsey and Hahn Echo ODMR sequences, respectively. The resulting values are T_2^*= 497 ns. and T_2= 840 ns.

These phase coherence times could be improved by the use of advanced dynamic decoupling techniques, including Carr-Purcell-Meiboom-Gill sequences, which have been demonstrated to provide an increase of a factor of two over spin echo in nanodiamond systems [6] and an order of magnitude in C^{13} nuclear-spin limited systems [8]. In addition, a reduction in nitrogen implant dose would further lower the number of paramagnetic impurities and boost coherence time. The NV density would be lowered as a consequence, but this could be counteracted by increasing implantation
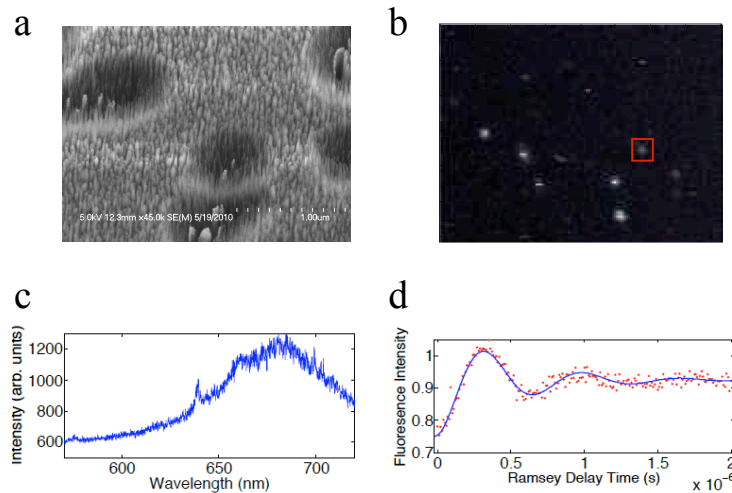
Fig. 1. (a) Scanning electron micrograph of CVD diamond nanocrystals attached to bulk diamond substrate. (b) Confocal fluorescence scan of detached CVD nanocrystals on a glass substrate. The red box indicates a nanocrystal containing a single quantum emitter as determined by the second order autocorrelation function. (c) Fluorescent spectrum of a detached nanocrystal, confirming the presence of NV with the characteristic zero-phonon line at 638 nm. (d) Ramsey measurement on a single NV showing spin phase coherence time T_2^* = 497 ns.

yield, for example through use of a C^{12} co-implant [9]. Even without these optimizations, our spin system sets a record T_2^* time for diamond nanocrystals, which underscores the viability of this method for creation of long-coherence NV systems. When combined with the ease of mass parallel fabrication, these CVD diamond nanocrystals can enable a multitude of sensing and quantum information procedures at the nanoscale.

References

1. I. Aharonovich and A. Greentree, "Diamond photonics," Nature Photonics **5**, 397–405 (2011).
2. J. M. Taylor, P. Cappellaro, L. Childress, L. Jiang, D. Budker, P. R. Hemmer, a. Yacoby, R. Walsworth, and M. D. Lukin, "High-sensitivity diamond magnetometer with nanoscale resolution," Nature Physics **4**, 810–816 (2008).
3. F. Dolde, H. Fedder, M. W. Doherty, T. Nöbauer, F. Rempp, G. Balasubramanian, T. Wolf, F. Reinhard, L. C. L. Hollenberg, F. Jelezko, and J. Wrachtrup, "Electric-field sensing using single diamond spins," Nature Physics **7**, 459–463 (2011).
4. E. Rittweger, K. Y. Han, S. E. Irvine, C. Eggeling, and S. W. Hell, "STED microscopy reveals crystal colour centres with nanometric resolution," **3**, 1–4 (2009).
5. E. H. Chen, O. Gaathon, M. E. Trusheim, and D. Englund, "Wide-field multispectral super-resolution imaging using spin-dependent fluorescence in nanodiamonds," Submitted (2012).
6. A. Laraoui, J. S. Hodges, and C. a. Meriles, "Nitrogen-Vacancy-Assisted Magnetometry of Paramagnetic Centers in an Individual Diamond Nanocrystal." Nano letters (2012).
7. M. J. Burek, N. P. de Leon, B. J. Shields, B. J. M. Hausmann, Y. Chu, Q. Quan, A. S. Zibrov, H. Park, M. D. Lukin, and M. Lončar, "Free-standing mechanical and photonic nanostructures in single-crystal diamond." Nano letters **12**, 6084–9 (2012).
8. B. Naydenov, F. Dolde, L. Hall, C. Shin, H. Fedder, L. Hollenberg, F. Jelezko, and J. Wrachtrup, "Dynamical decoupling of a single-electron spin at room temperature," Physical Review B **83**, 1–4 (2011).
9. J. Schwartz, P. Michaelides, C. D. Weis, and T. Schenkel, "In situ optimization of co-implantation and substrate temperature conditions for nitrogen-vacancy center formation in single-crystal diamonds," New Journal of Physics **13**, 035,022 (2011).