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Abstract
We report on wide-field optically detected magnetic resonance imaging of nitrogen-vacancy centers (NVs) in type IIa polycrystalline diamond. These studies reveal a heterogeneous crystalline environment that produces a varied density of NV centers, including preferential orientation within some individual crystal grains, but preserves long spin coherence times. Using the native NVs as nanoscale sensors, we introduce a three-dimensional strain imaging technique with high sensitivity ($< 10^{-5}$ Hz$^{-1/2}$) and diffraction-limited resolution across a wide field of view.

1. Introduction
Recent years have seen rapid advances in the development of quantum memories and sensors based on solid-state spin systems. At the forefront of these spin systems is the nitrogen vacancy center in diamond (NV), which has an electron spin triplet ground state with exceptionally long coherence time even at room temperature [1]. Measuring these spins by optically detected magnetic resonance (ODMR) has enabled high performance sensing of electromagnetic fields [2, 3], temperature [4] and pressure [5]. The highest-sensitivity experiments have used single-crystal diamond grown through chemical vapor deposition. However, such samples remain expensive and small (on the scale of millimeters), which limits the adoption and scaling of NV sensing techniques. Polycrystalline diamond (PCD) presents an attractive alternative as it can be grown on the wafer scale and at lower cost, with the potential for long spin coherence times [6, 7]. However, a major concern with PCD has been that the diverse crystal structure, including grain boundaries and varied growth regimes, could lead to inconsistent and poor NV properties. In this work, we employ wide-field ODMR spectroscopy [8–11] to characterize the properties of hundreds of individually resolved NVs across a field of view of $> 300 \mu m^2$ and to use those centers as high-resolution nanoscale sensors of local crystal strain. These studies reveal that NVs in PCD are naturally preferentially aligned in some crystal grains, have spatially varying concentration, can be strongly strained near grain boundaries, and exhibit consistently long spin coherence times. Using these NVs, we introduce a method for wide-field strain imaging. We produce detailed, three-dimensional maps of PCD structure with diffraction-limited resolution and sensitivity below $10^{-5}$ Hz$^{-1/2}$, outperforming traditional optical strain measurement techniques such as Raman imaging [12]. These studies show the application of NVs for high-resolution strain imaging, and demonstrate the viability and potential advantages of PCD for quantum sensing and information processing.

The NV is an electronic spin-1 system consisting of a substitutional nitrogen atom adjacent to a vacancy in the diamond lattice [13]. The ground-state spin can be coherently manipulated by microwave fields, as well as initialized and detected through optical illumination because the $m_s = \pm 1$ sublevels are dark states that emit reduced fluorescence. The ground state Hamiltonian describing the system is:

$$H = \frac{1}{\hbar^2}[(D_{gs} + \epsilon_s)S_z^2 - \epsilon_s(S_x^2 - S_y^2) + \epsilon_x(S_xS_y + S_yS_z)] + \frac{gI\mu_B}{\hbar} \mathbf{S} \cdot \mathbf{B},$$

where $D_{gs} = 2.87$ GHz is the spin–spin interaction energy, $\epsilon = d_{gs} \cdot (E + \mathbf{\sigma})$ is the energy of interaction with external electric (E) and strain ($\mathbf{\sigma}$) fields, B is the magnetic field at the location of the NV, $d_{gs}$ is the NV electric
To probe the properties of NVs in PCD, we performed wide-field ODMR measurements using a custom-built fluorescence microscope (figure 1(a)). Optical illumination is provided by a 532 nm green laser modulated by a double-pass acousto-optic modulator, and resulting NV fluorescence is spectrally filtered (650 nm long-pass) and collected on an electron-multiplying CCD camera. Spin manipulation is accomplished by applying microwaves through a 15 μm copper wire placed nearby to the area of interest, while the external magnetic field is controlled by three orthogonal current-controlled electromagnets in addition to a permanent rare-earth magnet. The measurements were performed at room temperature without external stabilization or control.

We investigated a type-IIa PCD provided by Element6, grown by chemical vapor deposition with a nitrogen concentration <50 ppb. We first imaged the sample in fluorescence. Individual crystal grains vary in size from 10–1000 μm² in area, and are easily identified in fluorescence imaging by their boundaries (figures 1(c) and (d)). These grain boundaries fluoresce bright across the visible band, likely due to a high density of optically active lattice traps and amorphous carbon [7], and indicate the transition between two growth regimes. Individual NV centers within individual PCD grains are visible under 100 x magnification (figures 1(b) and (c)). The measurements show an NV density that varies significantly within and between grains, from roughly ~0.1
NV $\mu m^{-2}$ to densities approaching 1 NV $\mu m^{-2}$. This heterogeneity contrasts with the homogenous NV distribution in single-crystal CVD diamond.

We then investigated NV properties in the high magnetic field regime. An external magnetic field of $\sim 100$ G was applied and the ODMR spectrum of NVs within the field of view obtained under continuous-wave microwave and optical excitation (figures 2(a) and (b)). The observed resonance frequencies correspond to specific geometric classes of NVs \{i\}, each with a defined angle to the external magnetic field resulting in a different resonance frequency $\hbar \omega_i = D_{gs} + g\mu_B B \cos \theta_i$. Interestingly, only three distinct NV orientations appear in this region, rather than the four possible classes corresponding the four crystallographic (111) directions. In figures 2(c) and (d), we map the location of NV geometric classes \{i = 1, 2\} by showing ODMR at frequencies $\omega_i$. This shows spatial separation of NV classes, likely by growth region as no intersecting grain boundary is observed, with the lower region only containing NVs at a single frequency. This effect persists independent of the direction of the applied permanent magnetic field (supplemental material), and across grains in the PCD. Preferential orientation of NVs through engineered growth has been theoretically predicted \[16, 17\] and observed previously in single crystal diamond with controlled growth along \{110\} \[18, 19\], \{111\} \[20, 21\] and \{113\} \[22\]. These results indicate that preferential alignment occurs naturally in PCD, possibly due to predominant \{110\} and \{111\} grain textures.

3. Strain imaging

We next turn to strain mapping of the PCD by wide-field continuous-wave ODMR spectroscopy. These measurements must be performed in the low magnetic field regime where $B_z \ll E_z$; to achieve this, we canceled external magnetic fields using three-axis electromagnets. Figure 3(b) shows a representative ODMR spectrum. From double-Lorentzian fits to these low-field spectra, taken in parallel across the field of view with a total measurement time of 150 s, we extracted $D_{gs}$ and $E_z$, following equation (2), and converted them to strain. The resulting strain maps are shown in figures 3(c) and (d), respectively. Axial strain values relative to a $D_{gs}$ parameter of 2.87 GHz are shown. For non-axial strain, the minimum detectable resonance splitting is set by the magnetic field induced from the NV-site $^{14}$N nuclear spin \[3\], allowing for absolute calibration of a zero value. The possible presence of multiple NV orientations could lead to a worst-case underestimation of strain through both the axial and non-axial parameters by a factor of two. Figure 3(c) indicates a maximal axial strain of $6 \times 10^{-4}$ at the grain boundary which relaxes towards the center of the grain, while figure 3(d) indicates a...
maximum non-axial strain of $1.8 \times 10^{-4}$. These strains relax to their minimum value over a distance of 24 $\mu$m from the grain boundary, setting a relevant length scale of the use of PCD in device design.

The sensitivity of this technique can be characterized by the 68% confidence interval on the fitted resonance frequencies, which corresponds to the measurement standard deviation and depends primarily on the detected photon rate. Due to the non-uniform illumination across the field of view as well as background fluorescence near grain boundaries, the sensitivity varies spatially. The mean 68% confidence interval on the two resonance frequencies for each 320 $\times$ 320 nm$^2$ pixel in the field of view is displayed in figure 4. We achieve a median per-pixel ODMR spectral resolution of 245 kHz, resulting in an axial strain measurement precision of $2.7 \times 10^{-5}$ (non-axial $1.2 \times 10^{-5}$) and a corresponding sensitivity of $3.21 \times 10^{-4}$ Hz$^{-1/2}$ (non-axial $1.5 \times 10^{-4}$ Hz$^{-1/2}$) for a 150 second measurement. To characterize a best-case sensitivity without the sample-specific background near the grain boundaries, we focus on the high-SNR regions corresponding to in-focus individual NVs at the center of the field of view. In these regions, we achieve 68% confidence intervals of 79 kHz, corresponding to a measurement precision of $8.2 \times 10^{-6}$ (non-axial $3.8 \times 10^{-6}$) and sensitivity of $1.02 \times 10^{-4}$ Hz$^{-1/2}$ (non-axial $4.7 \times 10^{-5}$ Hz$^{-1/2}$).
Using this low magnetic field ODMR technique, we performed wide-field strain imaging in three-dimensions across several crystal grains. These studies again reveal a strong strain gradient near grain boundaries. In the grain shown in figures 3(a)–(c), the $D_{\rho}$ parameter corresponding to axial strain is lower near the boundary than in the center of the grain. This corresponds to tensile rather than compressive strain, while the reverse is true for the grain shown in figures 3(c) and (d). The non-axial strain at the center of the field of view is reduced with increased depth into the diamond, from a maximal value $>1 \times 10^{-4}$ near the surface to $<5 \times 10^{-5}$ at a depth of 3 $\mu$m. As visible in the fluorescence profile (figure 5(a)), the location and angle of the grain boundary varies with depth into the diamond, which is in turn reflected by rotation in the axial strain profile (figures 5(b) and (c)). Here the strain is observed to relax within 10 $\mu$m from the grain boundary. Further strain images, including extreme strain gradients and comparisons to reference single-crystal diamonds, are included in the supplemental material. These maps demonstrate the power of this technique for imaging crystal strain with high precision in three-dimensions.

4. Discussion

Our studies reveal advantages and disadvantages of PCD for NV-based applications. The observation of preferential NV alignment within PCD grains, in combination with consistently high NV spin coherence times similar to single-crystal samples, can greatly improve the signal-to-noise of NV sensing applications (i.e. a fourfold improvement in ODMR contrast) [18]; the large areas available for PCD diamond add to the usefulness in wide-field sensing applications, for example in biology [23]. Strong strain gradients could also allow for sub-diffraction resolution of individual NVs using ODMR [11]. In photonic devices, well-defined angular alignment of NVs can improve NV-dipole coupling to optical modes. PCD enables the production of wafer-scale photonic circuits in diamond, but roughness and scattering from grain boundaries have limited device performance compared to single-crystal diamond [24]. Large grains with low intra-grain roughness, such as those present in the sample characterized in this work, may reduce loss while still providing large-area substrates. Wide-field pre-characterization of the PCD could also allow for identification of, and design compensation for, grain boundaries.

PCD could also serve as a natural environment for studying correlations between crystal strain and the spin and optical properties of embedded emitters. For example, high strain could break the orbital degeneracy of the SiV [25], changing spin and orbital coherence properties, or strain gradients could be employed to tune relative NV sensitivity to electric and magnetic fields [26]. PCD provides a substrate that is ultra-pure and enables long spin coherence times, in contrast to other highly strained host materials such as nanodiamond. The strain in PCD can be very large; the observed NV ground state spin strain shifts of over 8MHz in the axial direction are higher than those reported in cantilever-based experiments [14, 15] and would require pressures in the tens of GPa to produce externally.

5. Conclusion and outlook

The NV-based strain imaging technique introduced in this work reaches the optical diffraction limit and a high sensitivity of $10^{-5}$ Hz$^{-1/2}$ ($<10$ MPa), which outperforms more traditional strain imaging techniques such as Raman imaging [12] that are limited to absolute sensitivities $>10$ MPa in addition to being orders of magnitude slower [27]. Through sectional imaging, this 3D imaging technique also offers an advantage over birefringence.
[28, 29] or x-ray topography [30] strain imaging methods, which image whole-sample and near-surface strains, respectively. Although limited to diamond and other materials with optically accessible, strain-sensitive spin defects (e.g. silicon carbide [31]), this technique has potential for precisely characterizing internal strains, such as those induced by geological formation [32] or in strain-engineered devices, as well as for mapping externally applied strains. More advanced dynamical-decoupling sequences can increase axial strain sensitivity [33, 34], which in turn could enable sub-diffraction strain imaging by resolving NVs in the spectral domain. While this work images individual NVs, higher-NV-density samples would increase sensitivity [35], potentially enabling the imaging of few-site dislocations in single-crystal diamond or increasing the local strain. Since the NV is a truly nanoscale sensor, operable over a wide range of temperatures [36] and pressures [5], this method can be used to perform ultra-high resolution strain mappings dynamically in previously unexplored regimes.

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