Imaging Strain and Electric Fields in NV Ensembles using Stark Shift Measurements

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We report measurements of optically detected magnetic resonance spectra of ensembles of negatively charged nitrogen-vacancy (NV) centers in diamonds in the presence of strain and DC external electric fields. The Stark shift of the spectral lines is stronger when the axial magnetic field along the NV center’s quantization axis is minimized. The shift is also enhanced at avoided crossings between the hyperfine levels at an axial field of $B = \pm 77 \mu T$. Since the intrinsic strain in the diamond also induces a Stark shift, we are able to calculate the magnitude and direction of the strain within the crystal. We also use the Stark effect to map the electric field in the diamond volume between patterned electrodes.

I. INTRODUCTION

Nitrogen-vacancy centers are formed in diamonds when a nitrogen atom substitutes for one of the carbon atoms in the lattice and it pairs with a vacancy. A center can acquire an electron to form NV$^-$, which we hearafter indicate as NV. The defect center can also be electrically neutral (NV$^0$) or carry a positive charge as the optically inactive (NV$^+$).1

NV centers have relatively long-lived spin coherence features that can be initialized and detected with visible light.2 Recent studies of single NV centers are motivated by applications including quantum computing3 and nanoscale sensing of magnetic4 and electric5 fields.

The electronic ground state $^3A_2$ is an orbital singlet state and consists of a $m_s = 0$ level separated from nearly degenerate $m_s = \pm 1$ levels by $D_{gs} \approx 2.87$ GHz (Fig. 1). The NV center can absorb off-resonant 532 nm light, after which it fluoresces from the $^3E$ excited state. The zero phonon line (ZPL) is 637 nm, but significant fluorescence is observed in wavelengths up to 800 nm. A competing non-radiative pathway via intermediate singlet states also exists for the excited state and is stronger for the excited state $m_s = \pm 1$ levels. Because of this non-radiative pathway, the overall fluorescence of the excited state depends on the excited state $m_s = \pm 1$ population which in turn depends on the ground state $m_s = \pm 1$ population. This allows the spin of the ground state of the NV center to be determined by monitoring the brightness of the emitted fluorescence.2

Either CW or pulsed microwave techniques can be used to examine magnetic dipole transitions from the $m_s = 0$ to $m_s = \pm 1$ levels in the ground state with high spectral resolution.2 This technique of resolving the ground state electronic structure is called optically detected magnetic resonance (ODMR) and we use it in this work to measure DC Stark shifts.

Recent work5 has shown how a single NV can be used for measuring the transverse component of the DC electric field and how NV ensembles can be used to measure electric fields applied across a 300 um wide diamond.7

We are interested in developing the capability to measure electric and magnetic fields near the interaction region of experiments to measure the electric dipole moment of the neutron.8 Such experiments seek to push the boundaries of the Standard Model. It is critical that the probe makes only small perturbations to the fields that it is measuring. In this regard, NV diamonds show promise, as the diamond is electrically and magnetically inert. While this work uses microwaves, previous work has shown that magnetic9,10 and electric field11 effects can be detected with all-optical NV diamond probes. In this paper, we present measurements of strain and external electric fields using ODMR in NV ensembles.

To compare our results with theory, we model the ground state Hamiltonian, $\hat{H}_{gs}$ as

$$\hat{H}_{gs} = \hat{H}_{hfs} + \hat{H}_{es}.$$  

Here $\hat{H}_{hfs}$ is the zero-field Hamiltonian which includes the hyperfine interactions with the $^{14}$N (spin = 1) nucleus, $\hat{H}_{es}$ is the electronic spin Hamiltonian which includes the strain, magnetic and electric field interactions.

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FIG. 1. a) Structure of an NV center in the diamond lattice. The quantization axis of the NV center is along the line connecting the nitrogen atom and the vacancy. b) Relevant spectroscopy of NV centers, including ground state fine structure (not to scale). Only a single arm of the excited state doublet is shown. Hyperfine structure is also omitted for simplicity.

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The g-factor tensor, $\bar{g}$, includes longitudinal and transverse components.

The electronic spin component of the ground state energy is given by

$$\hat{H}_{eS} = \frac{1}{\hbar^2} d_{\parallel} \Pi_{z} S_z^2 + \frac{\mu B}{\hbar} \vec{S} \vec{B} - \frac{1}{\hbar^2} d_{\perp} \Pi_y (S_x^2 - S_y^2) - \frac{1}{\hbar^2} d_{\perp} \Pi_x (S_x S_y + S_y S_x).$$

Here, $\Pi = \vec{E} + \vec{\sigma}$ is the total effective electric field which also includes contributions from the strain, $\vec{\sigma}$. The ground state electric dipole moment, $d$, has a much larger transverse component $d_{\perp} \approx 17$ Hz cm/V compared to the longitudinal component, $d_{\parallel} \approx 0.3$ Hz cm/V. The effective g-factor tensor, $\vec{g}$ has a longitudinal, $g_{||}$ and a transverse, $g_{\perp}$ component as detailed in Ref. 6.

**II. EXPERIMENTAL DETAILS**

Diamonds formed by chemical vapor deposition with a nominal nitrogen content of about 100 ppb were obtained from Element 6. The diamonds are approximately 5x5x0.5 mm, cut so that the 5x5 mm$^2$ faces are (100) planes and the 0.5x5 mm$^2$ edges are (011) planes, as shown in Fig. 2. The diamonds were irradiated with a low-energy electron beam with an energy of 2 MeV and a fluence of 10$^{17}$ cm$^{-2}$ to generate the vacancies. They were then annealed for 2 hours at 875 K to encourage the coupling of vacancies with nitrogen nuclei. By integrating the visible absorption spectrum of the diamonds, the density of NV states after processing was estimated to be 20 ppb. Two chromium electrodes, a few hundred nanometers in thickness, were photolithographically deposited on the top surface of one diamond. The electrodes are separated by a gap that is about 80 um wide at the center of the surface, growing to about 350 um near the edges. The patterned electrodes create a slowly varying electric field in the diamond.

The experimental apparatus is shown schematically in Fig. 3. A specially constructed confocal microscope is used to investigate the ground state structure of NV ensembles. Green light from a frequency-doubled Nd:YAG laser can be adjusted in power with a variable attenuator, while the angle of the linear polarization can be adjusted with a half-wave plate. The light beam is brought onto the optical axis of a long working distance, 50x microscope objective by means of a dichroic mirror. The diamond is mounted in vacuum in a cryostat, with one electrode grounded to the cryostat as the potential of the other is varied from 0 to ±3000 V DC. The rear, (100) face of the diamond is also grounded. The light is focused into the gap between the electrodes using the microscope objective located outside the cryostat. Fluorescence is collimated by the objective and filtered through the dichroic beam combiner and by an optical longpass filter (Andover 590FG05, passing $\lambda > 600$ nm).
light is spatially filtered with a 50 um pinhole before being sent to an avalanche photodiode. The microwave source is a digitally-synthesized oscillator (WindFreak Synth NV) amplified to 25 dBm. The microwaves are coupled to the diamond using a resonant loop of wire, about 1 cm from the diamond, located outside the cryostat.

Since there are four possible crystallographic orientations of NV axes, we see four different sets of resonances from within the ensemble. The magnetic dipole transitions have the following selection rules: \( \Delta m_s = \pm 1 \) and \( \Delta m_I = 0 \); this leads to 6 ground state resonances. A total of 24 resonances (= 4 crystallographic orientations \( \times 6 \) hyperfine transitions) can be observed in the ODMR spectrum. Polarization of the green laser controls coupling strength among the four NV orientations. This technique allows the selective excitation of NV centers, as shown in Fig. 2.

III. RESULTS

The axial resolution is measured by moving the diamond through the focus of the microscope objective. As shown in Fig. 4a, the axial resolution is about 80 um. The transverse resolution, measured using a similar method, is about 1 um. By measuring the fluorescence intensity at the NV\(^{-} \) (637 nm) and NV\(^{0} \) (575 nm) ZPL, we can measure changes in their relative concentrations. Closer to the surface, in regions of large electric field \( (E > 40 \text{ kV/cm}) \), there was marked decrease in the concentration of NV\(^{0} \) centers with a simultaneous increase in NV\(^{-} \) centers (up to 100%). This effect was attenuated as we moved towards the grounded face of the diamond. We believe this effect is related to the charge state conversion observed in single NV centers because of field induced band-bending.

As shown in Fig. 4b, the difference in fluorescence at 0 V (blue), 2500 V (red) and their difference (yellow) represent the intensity of the 637 nm ZPL. Curves are rescaled for clarity. The red curves show the strain measured as a function of depth in the diamond. The difference in fluorescence at 0 V (blue), 2500 V (red) and their difference (yellow) show the strain direction as determined by the polar plots of the Stark shift is drawn in orange. The orange vector represents the strain as it projects on the transverse plane and the shaded region represents the uncertainty associated with the strain measurementw. Simulated data is shown as a blue curve.
is important to characterize the strain magnitude and direction for electric field measurements. As shown in Fig. 4a, the axial variation in the strain magnitude was measured by scanning the confocal microscope in the (100) crystallographic direction.

In order to measure the strain direction, the strain induced Stark shift was calculated for NVs along all four crystallographic directions. A largely orthogonal magnetic field with a small axial component of 77 uT was applied to isolate NV centers oriented along the four different (111) crystal directions. The magnitude of the strain was measured to be equal for all four axes. This narrowed down the strain heading to the three possible degenerate directions - (100), (010) and (001). Given this result, there is also the possibility of the strain to be isotropic.

To complete our strain analysis, we used the dependence of the magnetic field polar angle, $\theta_B$ in eq. (4) for a suitably large orthogonal magnetic field (1 mT), generated by a pair of Helmholtz coils and ceramic magnets. As the magnetic field was rotated in the non-axial plane, changes in the strain induced Stark shift were recorded. The polar plot from this experiment is shown in Fig. 5. Fitting the graph to the energy shifts predicted by the Hamiltonian shows that the strain points in the (100) direction, out of the large face of the diamond. This direction is also parallel to the crystal growth direction. The total strain was determined to be $\sigma = 21 \pm 4$ kV/cm.

**B. Electric Field**

The ODMR transitions are shifted by the component of the electric field in the plane perpendicular to the NV axis. The Stark shift is typically small, a few Hz per kV/cm, but the shift is enhanced when the magnetic field is adjusted so that levels of the appropriate symmetry are nearly degenerate. As described in other work, the shift is enhanced in the transitions to the $|m_S = +1, m_I = 0\rangle$ and $|m_S = -1, m_I = 0\rangle$ states when the axial magnetic field is zero. From the Hamiltonian given by Doherty, we can also predict similar enhancements near $B_z = 77$ uT where the $|-1, -1\rangle$ and $|+1, -1\rangle$ levels cross. Likewise, the $|-1, +1\rangle$ and $|+1, +1\rangle$ levels cross at $B_z = -77$ uT. Stark shift enhancement at the avoided crossings of these hyperfine sublevels is discussed in our previous work.

The electrodes were patterned to produce variation in the electric field. A map of the electric field was produced by moving the focal spot axially into the diamond and measuring the Stark shift for the same voltage difference across the electrodes, as shown in Fig. 6. The electric field measurements were obtained by subtracting the strain-induced Stark shift from the total Stark shift ($\vec{\Pi} = \vec{E} + \vec{\sigma}$).

The largest contributor to the systematic error was the uncertainty in the electric dipole moment, $d_\perp = 17 \pm 3$ Hz cm/V. With just statistical errors, the typical uncertainty in the electric field measurement was $\sigma = 2$ kV/cm. After including systematic uncertainties, the cumulative error was 5 kV/cm. This shows that we can map electric fields throughout the whole volume of the diamond.

Future work will show how this technique can use NV center ensembles to serve as vector electrometers. The orthogonal component of the electric field from each of the four NV axes can be used to recreate the electric field. We also plan to investigate the influence of electric and magnetic fields on decoherence rates of the NV center.

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