

Second-order Stark shifts exceeding 10 GHz in electrically contacted SiV[−] centers in diamond

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Negatively charged silicon vacancy centers (SiV[−]) in diamond exhibit excellent optical properties and, below 100 mK, excellent spin coherence, making them promising candidates for quantum technologies. However, the strain-induced inhomogeneous distribution of optical transition frequencies poses a challenge for scalability. We demonstrate electrical tuning of the SiV[−] center zero-phonon lines using in-plane contacts to apply moderate electric fields up to 45 MV/m. The second-order Stark shift exceeds 10 GHz, which is of the same order of magnitude as the 15-GHz inhomogeneous distribution of SiV[−] observed in emitters embedded in optical nanostructures such as photonic crystal nanocavities. Analysis of individual SiV[−] centers shows significant variation in effective polarizabilities between defects indicating that the effective polarizability strongly depends on local parameters like strain. The observed effective polarizabilities are ≈ 3 –25 times larger than those of tin vacancy centers, which we attribute to valence band resonances that delocalize the e_u wave functions. Photoluminescence excitation measurements reveal that optical linewidths increase moderately with applied electric field strength. Our results demonstrate that large electrical Stark shifts can overcome the inhomogeneous distribution of transition frequencies, representing a significant step toward scalable SiV[−]-based quantum technologies such as quantum repeaters.

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I. INTRODUCTION

Many quantum technologies require long coherence times for their quantum memories and efficient optical interfaces to send information over long distances [1,2]. In this context, color centers in diamond are promising spin memories and spin-photon interfaces and have already been integrated into electrical and optical nanostructures [3,4]. In particular, the inversion-symmetric group-IV vacancies in diamond, including silicon, germanium, and tin vacancies, have excellent optical properties such as near-transform-limited zero-phonon lines [5], electron spin coherence times up to milliseconds [6,7], and nuclear spin coherence times exceeding seconds [8]. For silicon vacancy centers (SiV[−]), temperatures below 100 mK are required to suppress phonon-mediated decoherence enabling these coherence times. Other group-IV color centers can be operated at higher temperatures [7]. Due to local strain and Coulomb fields caused by other defects in the crystal, the emission wavelength can vary slightly between emitters, making emitted photons distinguishable. Specifically, the inhomogeneous distribution of optical emission frequencies lies in the range of 15 GHz for implanted SiV[−]

incorporated into nanostructures, even after careful annealing [9]. Overcoming this inhomogeneous distribution of optical emission frequencies is a practical challenge when building systems with multiple optically interfaced color centers.

Currently, different strategies are under investigation for making emitted photons indistinguishable.

(1) When converting distinguishable SiV[−] photons to telecom frequencies, the target wavelength can be precisely tuned by adjusting the frequency of the pump laser used in the nonlinear frequency conversion process individually for each emitter [4,10].

(2) Strain can change the optical transition frequency and can be applied locally using advanced nanostructures such as electrically tunable cantilevers [11–14].

(3) Electric fields can tune the optical emission frequency via the DC Stark effect, an approach that has already been demonstrated for nitrogen and tin vacancy centers [15–19], however not for SiV centers.

Here, we investigate Stark tuning of SiV[−] centers, which provides straightforward electrical tunability with simple electrode structures. Our results show that the effective polarizability is 3–25 times larger for silicon vacancies compared to tin vacancies [17–19]. With an electrode spacing of 7.6 μm , the applied electric fields remain well below those demonstrated without electrical breakdown in comparable diamond devices [18], leaving substantial room for larger Stark shifts. Despite the moderate fields, we demonstrate tuning of optical emission frequencies by multiple gigahertz, which

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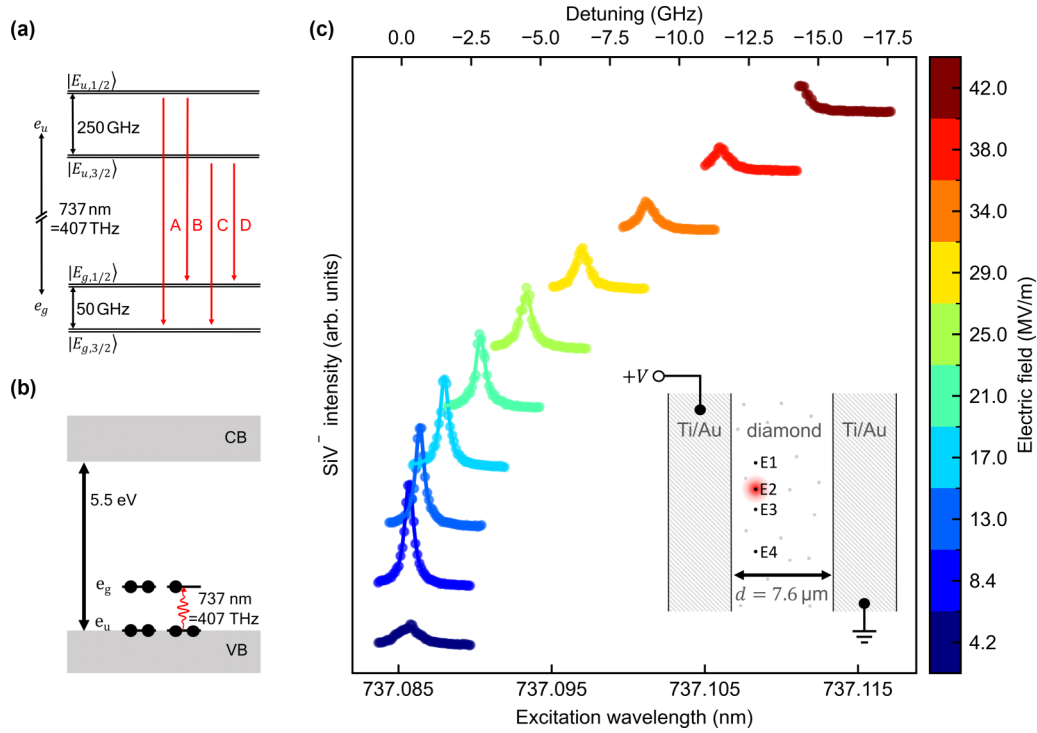


FIG. 1. SiV^- energy level diagram, sample schematic, and photoluminescence excitation spectra for varying applied electric fields. (a) Energy level diagram of SiV^- centers. The ground states $|E_{g,1/2}\rangle$ and $|E_{g,3/2}\rangle$, that have a hole in the e_g states, are split by approximately 50 GHz due to spin-orbit coupling and the Jahn-Teller effect. Strain can increase this splitting. The excited states $|E_{u,1/2}\rangle$ and $|E_{u,3/2}\rangle$, that both have a hole in the e_u orbitals, are lying close to the valence band maximum and split by at least 250 GHz. The red arrows define the optical transitions A to D that can be observed in photoluminescence and photoluminescence excitation. (b) Band diagram illustrating the multielectron configuration of the SiV^- ground state. (c) Inset: Top-view schematic of the relevant parts of the sample showing two thin Ti/Au electrodes on flat bulk diamond. The electrode distance is $7.6\ \mu\text{m}$ and we ground one of the electrodes and apply a voltage to the other. We focus the excitation and detection spot at approximately a quarter of the electrode distance from one of the electrodes. This breaks the symmetry in voltage because the photocurrent is stronger when the electrode closer to the excitation spot is positively biased. The small black spots E1, E2, E3, and E4 schematically mark different studied single SiV^- positions along the electrode edges. The gray spots mark other single SiV^- , which we did not select. (c) Photoluminescence excitation spectra of the emitter labeled E4, transition C (denoted E4-C), for different applied electric fields show that the optical emission frequency of SiV^- changes with the field. The solid lines represent Lorentzian peak fits of the data, individually performed for each voltage.

relaxes fabrication tolerances for addressing inhomogeneous broadening.

II. RESULTS

Figure 1(a) depicts the simplified energy level diagram of SiV^- in the absence of external magnetic fields [20,21]. The ground states $|E_{g,1/2}\rangle$ and $|E_{g,3/2}\rangle$ are split by approximately 50 GHz due to spin-orbit coupling and the Jahn-Teller effect, a splitting that can increase with strain. Figure 1(b) shows that these states are defined by the occupation of the energy levels e_u and e_g . The states e_u are close to the valence band maximum, while the energetically higher-lying states e_g are within the bandgap. In the ground states $|E_{g,1/2}\rangle$ and $|E_{g,3/2}\rangle$ of the SiV^- , the orbitals e_u are filled entirely with four electrons, while there are three electrons or effectively a single hole in e_g . For the excited states $|E_{u,1/2}\rangle$ and $|E_{u,3/2}\rangle$, the situation is reversed, meaning that there is a hole in the e_u levels [20]. The excited states are split by at least 250 GHz and their splitting can also increase with strain. The energy of the orbital states shifts with electric field as a consequence of the DC Stark effect [22]. Due to the inversion symmetry of the atomic

arrangement of a silicon vacancy complex [20,23], all SiV^- states are associated with inversion-symmetric wave functions. This leads to a vanishing static electric dipole moment and thus the absence of a first-order Stark shift. However, the negatively charged electron wave functions and positively charged nuclei can be polarized relative to each other by an applied electric field, leading to an induced electric dipole moment and a second-order shift of the electronic states' energies. Hence, no linear Stark shift is expected, but second-order Stark shifts can occur.

The Stark shift in the optical transition frequencies represents the difference of the Stark shifts of the participating initial and final states. We describe the optical transition frequency

$$f(E) = f_{\text{max}} - \alpha(E - E_0)^2 \quad (1)$$

and Stark shift

$$\Delta f = f(E) - f_{\text{max}} \quad (2)$$

as a function of an applied electric field E . Here, f_{max} is the maximum optical transition frequency, E is the applied electric field, E_0 is the local electric field offset, and α is the

effective polarizability of the transition frequency. The (total) effective polarizability α is the difference of polarizabilities of the two levels involved in the optical transition, where each of those is a measure of the field-induced perturbation of the wave function.

In this study, we use interdigitated electrodes on a flat oxygen-terminated diamond surface to apply an electric field to the silicon vacancies studied. The inset of Fig. 1(c) shows a schematic illustration of the electrodes and the measurement setting. The parallel electrode edges are separated by $7.6\ \mu\text{m}$. One of the electrodes is grounded, while the positive and negative contacts of the voltage source are floating to avoid ground loops. We position the focal spots from the excitation and detection channels at a quarter of the electrode distance. Since we keep this position distance constant, we have a constant conversion factor between applied voltage and applied electric field (see details below). For all measurements, we cool the sample to 5 K in vacuum (pressure $<10^{-5}$ mbar) using a closed-cycle cryostat.

To estimate the applied electric field experienced by the SiV centers for a given applied voltage, we simulate the system using the finite-element tool COMSOL and the Lorentz local field approximation. Details on the COMSOL simulation can be found in the Supplemental Material [24]. The Lorentz approximation accounts for the polarization of a medium surrounding a color center, which effectively enhances the electric field at the defect site [25]. The applied electric field acting on the defect is then given by $E = E_{\text{ext}}(\epsilon + 2)/3$, where E_{ext} is the externally applied field obtained from the COMSOL simulations and ϵ is the dielectric constant of diamond ($\epsilon = 5.7$) [18]. For an emitter 100 nm below the diamond surface and located $1.9\ \mu\text{m}$ (quarter of the electrode separation) from an electrode with an applied voltage of 10 V, the simulation gives $E_{\text{ext}} = 0.82\ \text{MVm}^{-1}$. This yields an applied field $E = 0.82\ \text{MVm}^{-1} \times (5.7 + 2)/3 = 2.10\ \text{MVm}^{-1}$. The uncertainty in the external field, based on assumed variations of the emitter position of 1.5 to $2.4\ \mu\text{m}$ from the electrode, translates to a 7% relative uncertainty in the applied field estimation. This sample has $\langle 110 \rangle$ edges. The SiV symmetry axes lie along $\langle 111 \rangle$ directions, of which two subtend an angle of 35.3° with the applied electric field and two are perpendicular to it. Due to the D_{3d} symmetry of the SiV center, the polarizability along the symmetry axis differs from that perpendicular to it, such that two families of SiV centers might be expected to exhibit different Stark tuning polarizabilities. In our experimental data, we do not observe two clearly distinguishable classes of data corresponding to these two orientation groups, rather a continuum of different polarizabilities indicative of the presence of inhomogeneous broadening.

To locate SiV⁻ centers, we translate the laser spot parallel to the electrode edge and measure the photoluminescence (PL) using a blue or green laser with either 740 THz/405 nm, 582 THz/515 nm or 564 THz/532 nm optical frequency and an optical power of 0.5–3 mW for excitation. The $1/e^2$ beam waist is approximately 600 nm. A representative PL spectrum is displayed in the Supplemental Material Fig. S1 [24]. For the given SiV⁻ density, we always find several SiV⁻ centers per detection volume, but they can be readily isolated spectrally by resonantly driving individual transitions. This means that the density is too high to resolve single SiV⁻ centers in PL

measurements, but low enough to distinguish them in resonant scans.

To strongly increase the spectral resolution and thereby effectively work with single SiV⁻ centers, we measure PL excitation spectra in the following. For this, we scan the optical frequency of a narrow-linewidth (<2 MHz) wavelength-stabilized laser over the peaks of the photoluminescence spectra around 737 nm. To stabilize the SiV in the negative state SiV⁻, we use one of two off-resonant low power green lasers with slightly different wavelength. For emitters E7 and E8 we used a blue laser instead. We used different lasers due to their limited availability. However, we confirmed experimentally that all three lasers are suitable to guarantee charge-state stability and thus refer to them simply as off-resonant lasers in the following. Details including the used laser and its power can be found in Supplemental Material Table S1 [24]. Both the resonant and off-resonant laser are continuous-wave and constantly on during the measurement. A 375- to 400-THz (750- to 800-nm) bandpass filter isolates the phonon sideband emission while suppressing the excitation lasers. To detect the emitted single photons we use single photon avalanche diodes or superconducting nanowire single photon detectors.

Figure 1(c) shows typical PL excitation spectra of an SiV⁻ center as a function of the applied electric field. We clearly observe a PL peak shift due to the DC Stark effect. Each spectrum is fitted using a Lorentzian to obtain the center frequency $f(E)$, peak amplitude, and peak linewidth as a function of the applied electric field. The resulting field-dependent data are presented in Fig. 2(a) confirming that the peak frequency shift Δf is quadratic in the applied voltage or electric field E . As discussed above, this is expected since SiV⁻ are inversion symmetric, leading to a vanishing linear contribution to the Stark shift. A second-order shift is also observed in the Stark tuning of tin vacancies (SnV⁻), which also exhibit inversion symmetry [17–19]. The data presented in Fig. 2(a) show two different SiV centers. Both show a quadratic Stark shift, but each clearly has a different effective polarizability. To reflect the possible modification of the externally applied electric field by optically induced electric fields, we denote the fit results as the *effective* polarizability α . One of the two emitters (E8-C) shifts by 4 GHz, while the other (E4-C) shifts by 13 GHz at the same applied electric field.

In an earlier work, we showed that a variation in the applied voltage changes the SiV charge state [28]. This process is induced by negatively biasing the electrode nearest to the SiV center, a procedure that promotes a transition to a dark state. Figure 2(b) shows the PL intensity as a function of the applied field, exhibiting similar behavior as observed in Ref. [28]: The SiV⁻ photoluminescence vanishes completely for very high positive or moderate negative bias voltages and we can no longer extract position and linewidth of the zero-phonon line from a photoluminescence excitation spectrum. The amplitude is highest for positive voltages between 50 and 100 V. We investigate the mechanism behind this charge conversion of individual SiV centers in much greater detail in a separate work, where we also show that SiV centers leaning to the SiV²⁻ charge state can be stabilized by a hole photocurrent [29].

We continue to discuss the linewidth as a function of the applied field. From the literature, we expect that the

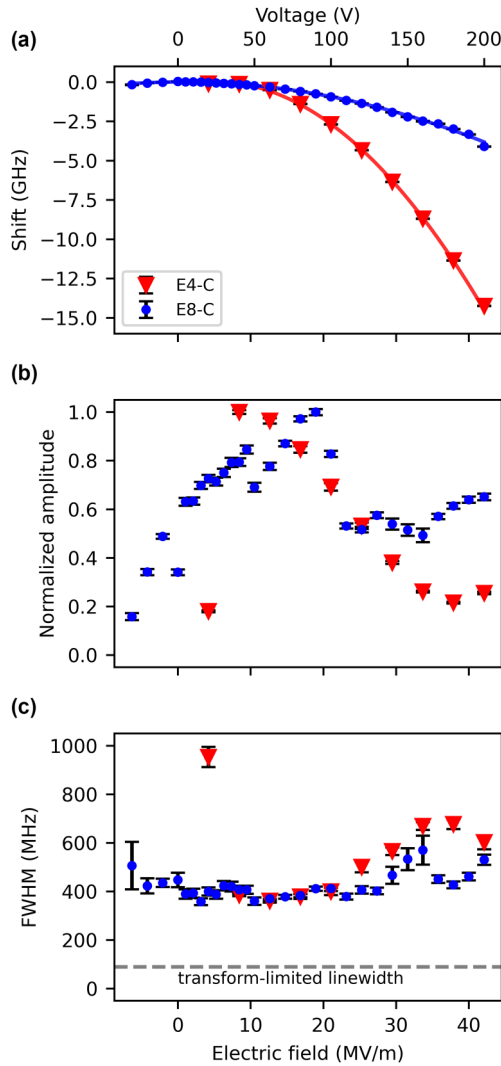


FIG. 2. Fitted peak shift, amplitude, and linewidth of an SiV^- zero-phonon line resonance as a function of the applied electric field for two different single SiV^- emitters. Shown are the data for emitters labeled E4 (red) and E8 (blue), transition C (denoted E4-C and E8-C). Error bars indicate the uncertainties given by the fit. (a) Peak center frequency shifts Δf with quadratic fit according to Eqs. (1) and (2). (b) Peak amplitudes. (c) Full width at half maximum linewidths compared to the literature value of the transform-limited full width at half maximum linewidth $1/(2\pi T_{1,\text{optical}}) \approx 90$ MHz [26,27].

transform-limited full width at half maximum (FWHM) linewidth is $1/(2\pi T_{1,\text{optical}}) = 90$ MHz [26,27]. Figure 2(c) shows the fitted FWHM linewidths as a function of the applied electric field. In the absence of an applied field, the linewidth is approximately 400 MHz, increasing to values between 500 and 600 MHz towards high applied fields. We expect that the additional line broadening is caused by a combination of the increasing induced static dipole moment that makes the center more sensitive to charge noise arising from current flow and fluctuating charge traps. Hence, the linewidth is expected to decrease by stabilizing the charge traps and lowering the photocurrent caused by the off-resonant laser

that stabilizes the SiV^- charge state, e.g., by using a pulse sequence. Additionally, the linewidth tends to increase with decreasing charge-state stability. The reason could be that when the charge state becomes unstable, the charge environment around the SiV is fluctuating more, causing electric field noise from nearby defects.

We measured the PL voltage-dependent excitation spectra for nine different emitters and observe center-to-center statistical variations of Stark tuning parameters. Figure 3(a) summarizes all peak center frequencies measured for transition C (defined in Fig. 1) of all centers and Fig. 3(b) shows the equivalent data for transition D. Supplemental Material Fig. S3 [24] shows a plot of all measured center frequencies plotted as a function of the applied electric field. We clearly observe a trend that the optical transition frequencies at zero applied field vary, resembling the typical inhomogeneous distribution caused by locally varying strain and electric fields induced by crystal defects [9]. The inhomogeneous distribution is <10 GHz for both transitions C and D. This is expected for annealed bulk diamond and somewhat smaller than the inhomogeneous distribution of around 15 GHz achieved for SiV^- in optical nanostructures [9].

Figure 3(c) shows the effective polarizability α as a function of the offset electric field E_0 . The measured SiV^- polarizabilities are significantly higher than those reported for tin vacancies [17–19] and, rather than falling into the two classes expected from the crystal symmetry, vary continuously between emitters. However, no direct correlation is observed between the effective polarizability and the zero-field frequency, i.e., the effective polarizability does not depend on the initial spectral position $f(E=0)$ of the resonance line. The offset electric field E_0 is always found to be positive and nonzero. This observation indicates that the spatial symmetry along the axis connecting the electrodes is broken. Furthermore, high polarizabilities occur preferentially alongside high local electric offset fields E_0 .

III. DISCUSSION

We observed center-to-center variations in the effective polarizability and the local electric field offsets E_0 . Band bending at the Schottky interface should be negligible compared to high applied voltages, considering that the whole barrier is limited to a few eV and that we apply up to 200 V. However, we attribute the relatively small different offset electric field E_0 to the charged surroundings of individual SiV centers. This includes the space-charge region caused by band bending at the nearby electrode and optical illumination [30], as well as the fluctuating charge environment caused by charged defects in the vicinity of each SiV^- [31]. Although the electrodes are nominally equivalent for all investigated positions, we find that the photocurrent depends on the exact location. Therefore, and because of the stochastic distribution of the defects, we do not expect E_0 to be perfectly homogeneous. After identifying an emitter, we typically performed a single measurement that included also negative voltages. However, the photoluminescence excitation peak intensity was consistently found to be very weak below 0 V or at least -20 V, consistent with our earlier work [28]. The exact voltage dependence varies between emitters. Consequently,

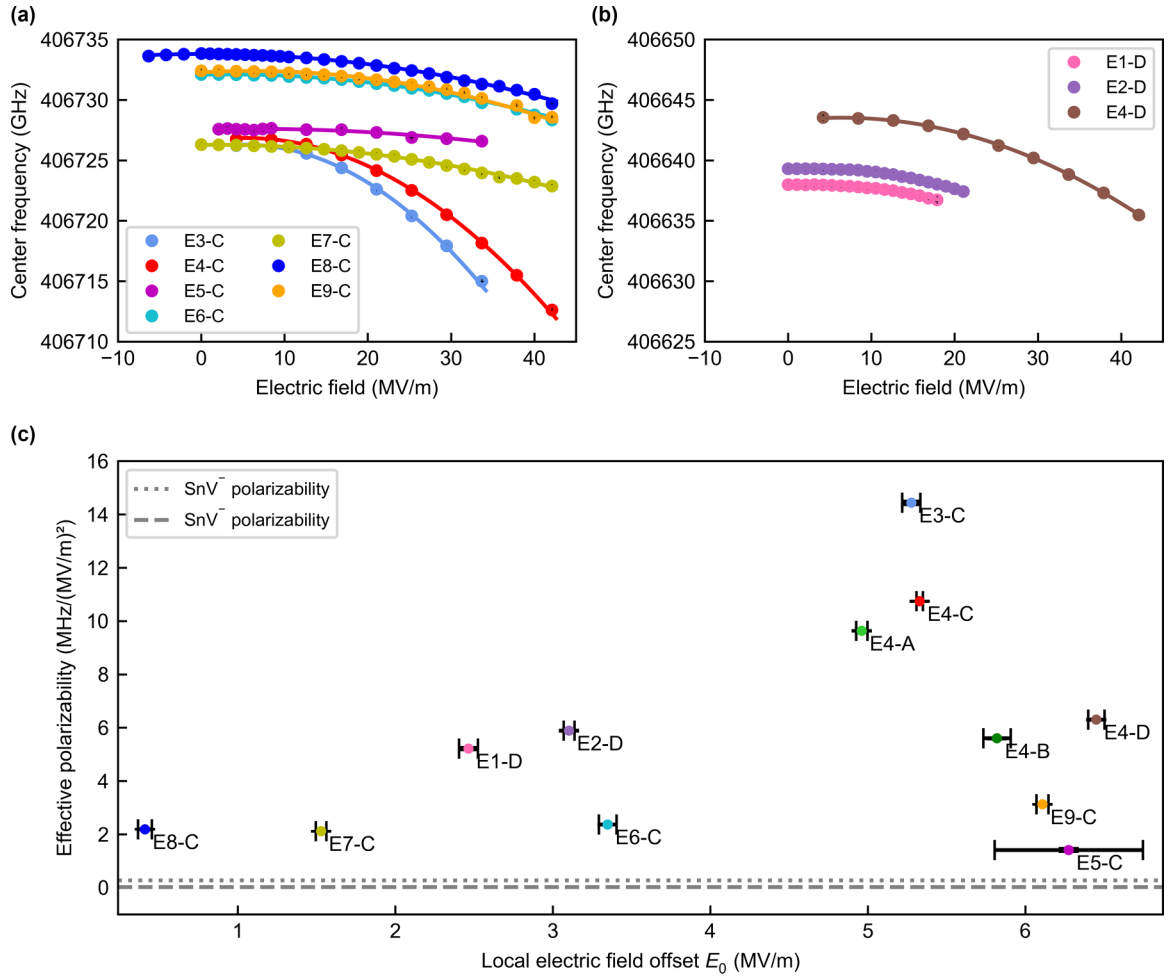


FIG. 3. Stark tuning of different individual emitters and statistics on analyzed parameters. The emitters are labeled from E1 to E9 and the measured transition is indicated with a letter from A to D as specified in Fig. 1(a). (a) Center frequencies of various emitters as a function of the applied electric field. Here, we show only the data that we measured for transition C. (b) Same as in (a) but for transitions D. (c) Correlation plot of fitted effective polarizabilities vs offset electric fields E_0 . The data indicate that high polarizabilities are accompanied by high local electric field offsets. Tin vacancy polarizabilities are shown for comparison [17,18].

we measured primarily using positive voltages, potentially leading to asymmetries in the charging behavior. This may also explain the exclusively positive values of E_0 , potentially since the optically induced space-charge region builds up slowly. Charged defects close to the SiV^- can only explain the symmetry breaking if they are a part of the optically induced space-charge region.

Two aspects can explain the strong variation in the observed polarizabilities between $1.4(1)$ and $15.0(1) \text{ MHz}/(\text{MV}/\text{m})^2$: First, locally varying properties such as strain and Coulomb fields from charged defects near the SiV^- can deform the wave function, hence altering the effective polarizability. Homogeneous strain preserves the local inversion symmetry while modifying the spatial extent of the wave functions, altering the polarizability without introducing a linear term. In this context, we note that only strain and Coulomb field *gradients* result in additional breaking of the inversion symmetry of the center, thereby resulting in a linear component to the observed Stark shift. It is known from literature that tin vacancies even show very strong linear contributions to the Stark shift, attributed to

strain [17]. However, we do not observe significant linear Stark shifts for our SiV^- emitters. Furthermore, strain may influence the response of NV^- centers to electric fields [32], a mechanism that could also translate to SiV^- centers. The lack of a clear correlation between effective polarizability and zero-field frequency $f(E=0)$ is complicated by the tensorial nature of both strain and the Stark response, whose principal axes need not necessarily coincide. Theoretical studies of the relationship between strain or nearby defects and effective polarizability could shed light on this.

Second, the variation of the polarizabilities may be an artifact arising from local electric fields that differ from the calculated values. This may arise due to screening of the applied electric field by an optically induced space charge region. Indeed, a study with a nitrogen-doped diamond found that space-charge regions built up during laser illumination and that they could screen the applied fields altogether at least up to a few MV/m [30]. Additionally, charges accumulating at the diamond surface can create orthogonal electric fields impacting the measured effective polarizability since the Stark tuning generally depends on the direction of the applied field

[33]. However, we do not expect the surface to influence the linewidths and charge states [5,26,34,35].

Our results indicate that large polarizabilities tend to occur alongside high offset electric fields E_0 . This correlation allows us to distinguish between two possible origins of the observed variation in polarizabilities. If the variation were purely an artifact of inhomogeneous screening, emitters experiencing stronger screening (and thus weaker effective fields) would appear to have higher polarizabilities. However, stronger screening also implies a stronger space-charge region and thus a higher E_0 . Consequently, the artifact hypothesis predicts an anticorrelation between E_0 and the apparent polarizability, the opposite of what we observe. The positive correlation in our data therefore supports the alternative hypothesis: that locally varying properties such as strain and Coulomb fields from nearby charged defects genuinely modify the electronic wave functions and hence the effective polarizability. Our arguments for the origin of E_0 , the spread of polarizabilities, and their relationship should serve as a first indication. Significantly more work will be needed to fully understand these effects. Further studies could measure all four optical transitions of each emitter, which would allow one to calculate the strain using the known Hamiltonian [11] and to directly correlate the measured strain to the effective polarizability.

Each Stark shift that we measured can be fitted well by Eq. (1), in contrast to a recent report where certain tin vacancies exhibited a very noticeable linear contribution to the Stark shift that was attributed to strong strain in the sample [17]. In addition to the absence of a linear term, we note that higher-order terms are not needed to fit the center frequencies, since the second-order fits describe the data very well. Data on the quadratic fits of all measured individual Stark shifts can be found in Supplemental Material Fig. S4 [24].

We attribute the substantially higher effective polarizability of SiV^- compared to SnV^- to differences in the confinement depths of the single-electron states. Density functional theory (DFT) calculations reveal that the e_u states of SiV^- are resonant with valence band maximum states and that they exhibit reduced localization [20]. This spatial delocalization enhances their size and effective polarizability and contributes to the larger Stark shifts observed in SiV^- centers. In contrast, the e_g states lie deep within the bandgap, resulting in strong localization and minimal contribution to the optical transition's Stark shift.

For heavier group-IV atoms, DFT calculations reveal that the e_u states move deeper within the bandgap [21], explaining the reduced effective polarizability observed in SnV^- centers. This systematic trend across group-IV vacancy centers provides fundamental insight into the relationship between electronic structure and characteristic electro-optical responses. An additional comparison can be made to the recently studied inversion-symmetric nickel vacancy in diamond [36]. The effective polarizability of this center is also significantly lower than that of the silicon vacancy and comparable to the effective polarizabilities of tin vacancies [17,18]. This is in agreement with our hypothesis above, as we do not expect one of the nickel vacancy energy levels to be resonant to the band edge states.

IV. CONCLUSION AND OUTLOOK

Our results demonstrate the potential for electrical tuning of SiV^- emission frequencies, for example to obtain indistinguishable photons for quantum repeaters. We find tuning ranges up to 15 GHz, which is in the same order of magnitude as the width of the inhomogeneous optical emission frequency distribution caused by strain [9]. We discuss the possibility that the SiV^- polarizabilities are higher compared to those of tin or nickel vacancies because the e_u level of the SiV^- is resonant to valence band maximum states and thus more delocalized, making it weakly confined and more polarizable. Furthermore, our measurements show the presence of offset local electric fields and provide insights about their origin and the observed variations in the effective polarizability between different emitters. We attribute the offset electric fields to an optically induced space charge region and the variations in effective polarizability to local perturbations like strain and Coulomb fields of other defects.

Turning to possible directions for future investigations, higher applied fields should significantly enhance the tuning range further and potentially allow frequency matching of arbitrarily selected SiV^- . Those higher fields could, e.g., be realized by electrodes with smaller separations of 1 to 4 μm . Significantly stronger electric fields could lead to charge-state instability, necessitating mitigation strategies for charge-state stabilization and charge-induced decoherence. Rapid voltage modulation protocols, as demonstrated in our previous work [28], could enable charge-state initialization before Stark tuning, potentially extending the accessible tuning range. Complete characterization of the Stark tensor through independent control of electric fields in all spatial dimensions would provide a comprehensive understanding of the electro-optical coupling. Finally, investigating the impact of rapidly varying electric fields on excited state dynamics may reveal new opportunities for photon shaping and quantum state manipulation.

V. MATERIALS AND METHODS

A. Sample preparation

Silicon vacancy centers were created by implanting silicon into electronic grade, single-crystal diamond produced by Element Six. Element Six grew the diamond using chemical vapor deposition and diced it into dimensions of $3 \times 3 \times 0.5 \text{ mm}$. This grade of diamond typically contains nitrogen and boron impurity levels below 5 and 0.5 ppb, respectively [37].

CuttingEdge Ions implanted ^{28}Si ions at 132 keV with an expected implantation depth of 100–150 nm and a dose of $5 \times 10^9 \text{ ions/cm}^2$. After implantation, the samples were annealed in multiple steps: 4-h ramp to 400 °C, 8 h at 400 °C, 12-h ramp to 800 °C, 8 h at 800 °C, 12-h ramp to 1100 °C, and 10 h at 1100 °C, all under high vacuum ($<1 \times 10^{-7} \text{ mbar}$) [9]. During this process, the temperature is gradually incremented to maintain high vacuum within the annealing chamber, essential for preventing surface graphitization of the diamond [38].

The sample was cleaned sequentially in aqua regia (1 h at 90 °C) and concentrated sulfuric acid (1 h at 225 °C), followed by oxygen plasma exposure to achieve oxygen termination. We fabricated interdigitated electrodes with 7.6 μm spacing using optical lithography and metal evaporation starting with 20 nm of titanium followed by 200 nm of gold. Each electrode connects to $200 \times 200 \mu\text{m}$ bond pads. Finally, we mounted the sample on a well-thermalized copper plate and wire-bonded it to a custom-printed circuit board chip carrier.

B. Optical and electrical measurements

The sample was cooled to 5 K using an attoDRY800 closed-cycle cryostat from attocube systems AG. Electrical voltage was applied using a Keithley 2450 source-measure unit, which also measured currents. Both the Keithley and half of the electrodes were grounded to the cold base plate of the cryostat in a star-shaped configuration.

Optical measurements were performed using a custom-built confocal microscope setup. Off-resonant excitation employed Toptica Photonics SE iBeam smart diode lasers with either 740 THz (405 nm) or 582 THz (515 nm) optical frequency or a torus laser from Laser Quantum LLC with an optical frequency of 564 THz (532 nm). The laser wavelengths and powers used for each measurement are shown in Supplemental Material Table S1 [24].

For resonant excitation, we used a narrow-linewidth continuous-wave laser (C-Wave GTR from Hübner Photonics), wavelength-stabilized to a wavemeter with 60-MHz absolute accuracy and 2-MHz resolution (High Finesse WS7-60 IR-I). Power stabilization was achieved using a software proportional-integral-derivative controller and analog modulation via a fast acousto-optic modulator (15-ns rise time, driven at 350 MHz in the first diffraction order).

All excitation lasers were coupled into single-mode fibers for spatial mode filtering. After combining both excitation lasers with a free-space dichroic mirror, a 400-THz (750-nm) frequency-long pass filter removed optical noise and fiber-

induced Raman signal frequencies from the excitation laser spectra. Laser focusing was accomplished using an $f = 2.87 \text{ mm}$, 0.82 NA, low-temperature apochromatic objective (LT-APO/VISIR/0.82 from attocube systems AG).

Detection employed single-mode fiber-coupled avalanche photodiodes with approximately 700-Hz dark counts or superconducting single-photon detectors from Single Quantum with $<1 \text{ Hz}$ dark counts. Three 400-THz (750-nm) frequency-short pass filters and one 545-THz (550-nm) frequency-short pass filter separated the excitation laser from the SiV^- phonon sideband signal. A 375-THz (800-nm) frequency-long pass filter removed the diamond Raman signal from the 407-Hz (737-nm) resonant excitation laser, which appears at 367 THz (816 nm).

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We used the artificial intelligence Claude from Anthropic PBC to review the understandability of our manuscript. Moreover, we acknowledge the use of ChatGPT and Claude for generating PYTHON code snippets used for experimental control software, as well as the analysis and processing of experimental data for publication.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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