

# Controlling the coherence of a diamond spin qubit through strain engineering

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The uncontrolled interaction of a quantum system with its environment is detrimental for quantum coherence. In the context of solid-state qubits, techniques to mitigate the impact of fluctuating electric [1–3] and magnetic fields [4–8] from the environment are well-developed. In contrast, suppression of decoherence from thermal lattice vibrations is typically achieved only by lowering the temperature of operation. Here, we use a nano-electro-mechanical system (NEMS) to mitigate the effect of thermal phonons on a solid-state quantum emitter without changing the system temperature. We study the silicon-vacancy (SiV) colour centre in diamond which has optical and spin transitions that are highly sensitive to phonons [9–13]. First, we show that its electronic orbitals are highly susceptible to local strain, leading to its high sensitivity to phonons. By controlling the strain environment, we manipulate the electronic levels of the emitter to probe, control, and eventually, suppress its interaction with the thermal phonon bath. Strain control allows for both an impressive range of optical tunability and significantly improved spin coherence. Finally, our findings indicate that it may be possible to achieve strong coupling between the SiV spin and single phonons, which can lead to the realisation of phonon-mediated quantum gates [14] and nonlinear quantum phononics [15–18].

Phonons couple to solid-state emitters directly through periodic deformation of the electronic wavefunctions [19]. Electron-phonon interactions are responsible for relaxation and decoherence

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32 processes in a variety of quantum systems [13, 20–25]. In particular, for systems with spin-orbit  
 33 coupling, phonon-mediated processes can demand operation at sub-Kelvin temperatures [26, 27], or  
 34 the use of magnetic fields of several Tesla [28] to achieve long spin relaxation and coherence times.  
 35 This requires cryogenic setups that are significantly more complex than common helium-4 cryostats  
 36 employed to obtain coherent optical photons from solid state emitters. In contrast, our approach  
 37 takes advantage of the fact that the large electron-phonon coupling responsible for such decoherence  
 38 processes fundamentally arises from a high susceptibility of the electronic orbitals to lattice strain.  
 39 We use this property to quench the effect of the thermal phonon bath on a single electronic  
 40 spin qubit without lowering the operating temperature. Our experiments are performed on the  
 41 negatively charged silicon-vacancy (SiV) centre in diamond, an emerging platform for photonic  
 42 quantum networks [29] with remarkable optical properties owing to its inversion symmetry [30].  
 43 This inversion symmetry is also responsible for the particular electronic structure of the SiV, shown  
 44 in Fig. 1a, with similar ground-state (GS) and excited-state (ES) manifolds, each containing two  
 45 distinct orbital branches [31]. Orbital degeneracy in each manifold is lifted by spin-orbit coupling:  
 46  $|1\rangle, |2\rangle$  in the GS split by 46 GHz, and  $|3\rangle, |4\rangle$  in the ES split by 255 GHz in the absence of strain.  
 47 Phonons with frequencies corresponding to these splittings can drive orbital transitions within the  
 48 ground and excited manifolds [13].

49 As a first step towards controlling the electron-phonon interaction, we investigate the effect  
 50 of static strain on these orbitals through strain-dependent photoluminescence excitation (PLE) of  
 51 the optical transitions labelled A, B, C and D at 4 K. Static strain control at the location of the  
 52 emitter is achieved with a NEMS device, a monolithic single-crystal diamond cantilever with metal  
 53 electrodes patterned above and below it [32], as shown in the scanning electron microscope (SEM)  
 54 image in Fig. 1b. An opening in the top electrode allows optical access to SiV centres located in  
 55 an array (inset of Fig. 1b), precisely positioned by focused ion-beam (FIB) implantation of  $^{28}\text{Si}^+$   
 56 ions [33, 34]. A DC voltage applied across the electrodes deflects the cantilever downwards due to  
 57 electrostatic attraction and generates controllable static strain oriented predominantly along the  
 58 long axis of the cantilever. The strain profile can be simulated numerically via the finite-element-  
 59 method (FEM), as shown in Fig. 1c. Of the two possible orientations of SiVs in our device, we  
 60 address those with transverse orientation (labelled blue, and shown in detail in inset of Fig. 1c),  
 61 which predominantly experience strain in the plane normal to their highest symmetry axis ( $E_g$  –  
 62 symmetric strain [35]). Upon applying strain, transitions A and D shift towards shorter and longer  
 63 wavelengths, respectively. These shifts indicate increasing GS and ES splittings as shown in Fig.  
 64 2a. This result is consistent with a previous experiment on a dense ensemble of SiVs [36]. The  
 65 variations in GS and ES splittings shown in Fig. 2a are quadratic at low strain, and linear at high

66 strain. This indicates that  $E_g$  – symmetric strain mixes orbitals within the GS and ES manifolds,  
 67 and thus phonon modes with corresponding strain components can induce resonant transitions  
 68 between these orbitals. In contrast, strain along the SiV axis ( $A_{1g}$  – symmetric strain) is found  
 69 to leave the GS and ES splittings unchanged, and therefore cannot cause electronic transitions.  
 70 Complete characterisation of the strain response and relevant group theory analysis are detailed  
 71 elsewhere [37].

72 With our device we can tune the splitting of the orbitals in the GS manifold from 46 GHz to  
 73 typically up to 500 GHz, and in the best case, up to 1.2 THz [32]. In doing so, we can probe the  
 74 interaction between SiV and the phonon bath at different frequencies by measuring the thermal  
 75 relaxation rate of the orbital with a time-resolved pump-probe technique (Fig. 2b). Measurements  
 76 are performed in the frequency range  $\Delta_{\text{gs}} = 46$  GHz to 110 GHz where this technique can be  
 77 applied. The total relaxation rate is a sum of the rates of phonon absorption,  $\gamma_{\text{up}}$ , and emission,  
 78  $\gamma_{\text{down}}$  (shown in Fig. 1a), which can be individually extracted using the theory described in [32].  
 79 Over the range of  $\Delta_{\text{gs}}$  measured, phonon processes in both directions are observed to accelerate  
 80 with increasing orbital splitting, thus indicating that the number of acoustic modes resonant with  
 81 the GS splitting, i.e. the phonon density of states (DOS) at this frequency, increases with an  
 82 expected dependence in  $\Delta_{\text{gs}}^n$  ( $n$  depends on the geometry of material seen by resonant phonons  
 83 [32]). However, if the orbital splitting is increased far above 120 GHz (at temperature  $T = 4$  K) as  
 84 plotted in Fig. 2c, the phonon absorption rate ( $\gamma_{\text{up}}$ ) is theoretically expected to reverse its initial  
 85 trend. In this regime, the polynomial increase in phonon DOS is outweighed by the exponentially  
 86 decrease in thermal phonon occupation ( $\sim \exp(-h\Delta_{\text{gs}}/k_B T)$ ) [13], and consequently  $\gamma_{\text{up}}$  is rapidly  
 87 quenched.

88 Such a suppression of phonon absorption at high strain can improve the spin coherence of the  
 89 emitter. In the presence of magnetic field, the SiV electronic levels further split into spin sub-  
 90 levels and provide an optically accessible spin qubit as shown in Fig. 3a [9, 10, 38]. We use  
 91 coherent population trapping (CPT) through simultaneous resonant laser excitation of the optical  
 92 transitions labeled C1 and C2 to pump the SiV into a dark state, a coherent superposition of  
 93 the spin sub-levels  $|1 \downarrow\rangle$ ,  $|1 \uparrow\rangle$ . When the two-photon detuning is scanned, preparation of the  
 94 dark state results in a fluorescence dip, whose linewidth is determined by the optical driving and  
 95 spin dephasing rates. At low laser powers, the linewidth is limited by spin dephasing, which is  
 96 dominated by phonon-mediated transitions within the GS manifold [13, 32]. In Fig. 3b, as the  
 97 dark resonance narrows down due to prolonged spin coherence with increasing strain, we reveal a  
 98 fine structure not visible before. Further measurements in Ref. [32] suggest that the presence of  
 99 two resonances is due to interaction of the SiV electron spin with a neighbouring spin such as a

100  $^{13}\text{C}$  nuclear spin. This indicates the possibility of achieving a local register of qubits as has been  
 101 demonstrated with nitrogen vacancy (NV) centres [39]. Fig. 3c shows the decreasing linewidths  
 102 of the CPT resonances with increasing GS orbital splitting, indicating an improved spin coherence  
 103 time. Beyond a GS splitting of  $\sim 400$  GHz, the linewidths saturate at  $\sim 1$  MHz. At the highest  
 104 strain condition, we perform a power dependent CPT measurement to eliminate the contribution  
 105 of power broadening, and extract a spin coherence time of  $T_2^* = 0.25 \pm 0.02 \mu\text{s}$  (compared with  
 106  $T_2^* = 40$  ns without strain control [9, 10]). This saturation of  $T_2^*$  suggests the mitigation of the  
 107 primary dephasing source, single-phonon transitions between the GS orbitals, and the emergence of  
 108 a secondary dephasing mechanism such as slowly varying magnetic fields from naturally abundant  
 109 (1.1%)  $^{13}\text{C}$  nuclear spins in diamond. Our longest  $T_2^* = 0.25 \pm 0.02 \mu\text{s}$  is on par with that of the  
 110 NV center without dynamical decoupling [4, 40], and of low-strain SiVs operated at a much lower  
 111 temperature of 100 mK [26], the conventional approach to suppress phonon-mediated dephasing.

112 In conclusion, we use a nano-electro-mechanical system to probe and control the interaction  
 113 between a single electronic spin and the phonon bath of its solid-state environment. In doing so, we  
 114 demonstrate six-fold prolongation of spin coherence by suppressing phonon-mediated dephasing as  
 115 the dominant decoherence mechanism. As a next step, we can further improve the spin coherence  
 116 by cancelling the effect of slowly-varying non-Markovian noise from the environment [26] using  
 117 dynamical decoupling techniques that are well-studied with other spin systems [6, 7, 39]. Our  
 118 strain engineering approach can be applied to overcome phonon-induced decoherence in other  
 119 emitters such as emerging inversion-symmetric centers in diamond [23, 24, 41, 42], Kramers rare  
 120 earth ions [25, 27, 28], and in general, systems with spin-orbit coupling in their ground state. High  
 121 strain needed to quench phonon processes can be achieved simply by deposition of a thin film [43],  
 122 which passively stresses the underlying crystal. A NEMS platform can provide the added benefit of  
 123 active wavelength tuning, which can enable generation of indistinguishable photons from multiple  
 124 emitters, and hence scalable photonic quantum networks [29, 44]. Another natural extension of our  
 125 work is coherent coupling of the SiV spin to a well-defined mechanical mode, which will enable the  
 126 use of phonons as quantum resource. In particular, we can combine the large strain susceptibility  
 127 of the SiV [37] with mechanical resonators of dimensions close to the phonon wavelength, such as  
 128 optomechanical crystals [45] to obtain orders of magnitude larger spin-phonon interaction strengths  
 129 compared with previous works [46–51], leading to strong spin-phonon coupling. In this regime, one  
 130 can realise phonon-mediated two-qubit gates [14] analogous to those implemented with trapped  
 131 ions [52], and achieve quantum non-linearities required to deterministically generate single phonons  
 132 and non-classical mechanical states [15–18, 53], a long sought-after goal since phonons can be used  
 133 to interface spins with other quantum systems such as superconducting qubits [54].

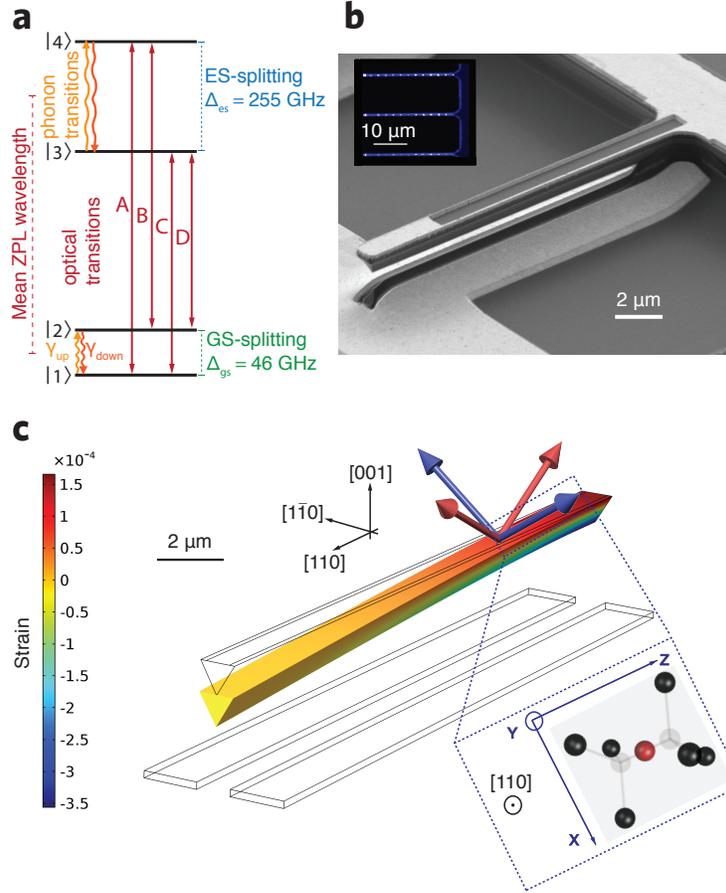


FIG. 1. (a) Electronic level structure of the SiV showing the mean zero phonon line (ZPL) wavelength, frequency splittings between orbital branches in the ground state (GS) and excited state (ES) ( $\Delta_{gs}$  and  $\Delta_{es}$  respectively) at zero strain, and the four optical transitions A, B, C, and D. Also shown are single-phonon transitions in the GS and ES manifolds. (b) Scanning electron microscope (SEM) image of a representative diamond NEMS cantilever. Dark regions correspond to diamond, and light regions correspond to metal electrodes. (Inset) Confocal photoluminescence image of three adjacent cantilevers. The array of bright spots in each cantilever is fluorescence from SiV centres. (c) Simulation of the displacement of the cantilever due to the application of a DC voltage of 200 V between the top and bottom electrodes. The component of the strain tensor along the long axis of the cantilever is displayed using the colour scale. Crystal axes of diamond are indicated in relation to the geometry of the cantilever. Arrows on top of the cantilever indicate the highest symmetry axes of four possible SiV orientations, and their colour indicates separation into two distinct classes upon application of strain. SiVs studied in this work are shown by blue arrows are oriented along  $[1\bar{1}1]$ ,  $[\bar{1}11]$  directions, are orthogonal to the cantilever long-axis, and experience strain predominantly in the plane normal to their highest symmetry axis. Inset shows the molecular structure of such a transverse orientation SiV along with its internal axes, when viewed in the plane normal to the  $[110]$  axis.

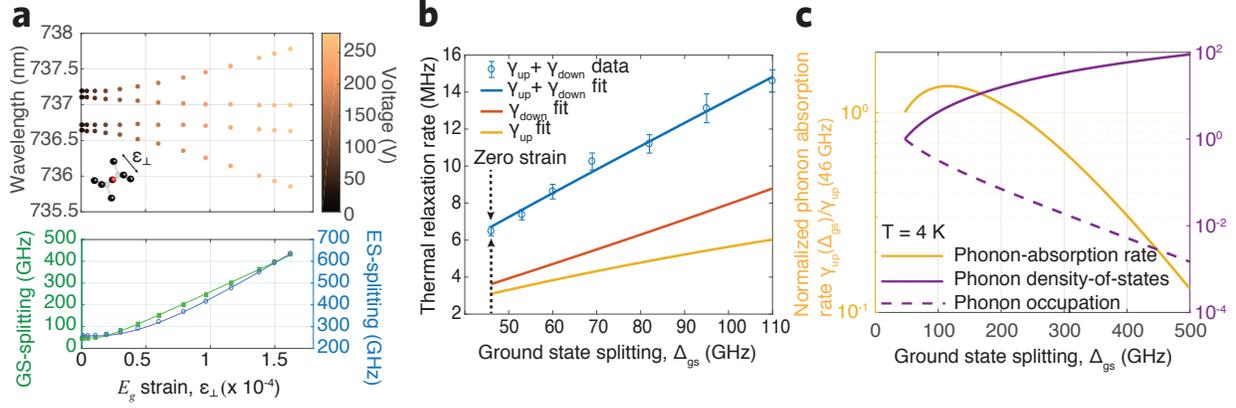


FIG. 2. (a) Strain response of a transverse orientation SiV labelled with a blue arrow in Fig. 1c. Wavelengths of the four optical transitions A, B, C, and D are recorded against strain. See [32] for raw PLE data with applied voltages. The lower panel shows orbital splittings within GS (solid green squares) and ES (open blue circles) extracted from the optical transition wavelengths. Solid curves are fits to group theory based strain response model [35, 37]. (b) Thermal relaxation rates between GS orbital branches vs. their energy splitting. Fit to model in [32] allows extraction of the phonon-absorption rate  $\gamma_{up}$  and phonon-emission rate  $\gamma_{down}$ . (c) Calculated phonon-absorption rate  $\gamma_{up}(\Delta_{gs})$  (solid yellow line) as a function of GS-orbital splitting  $\Delta_{gs}$  at temperature  $T = 4 \text{ K}$ . Left  $y$ -axis indicates the magnitude of this rate normalized to the value at zero strain,  $\gamma_{up}(46 \text{ GHz})$ . Right  $y$ -axis indicates the two competing factors whose product determines  $\gamma_{up}$ : the phonon density of states (normalized to its value at zero strain), shown with the solid violet line, and the thermal occupation of acoustic modes shown with the dashed violet line.

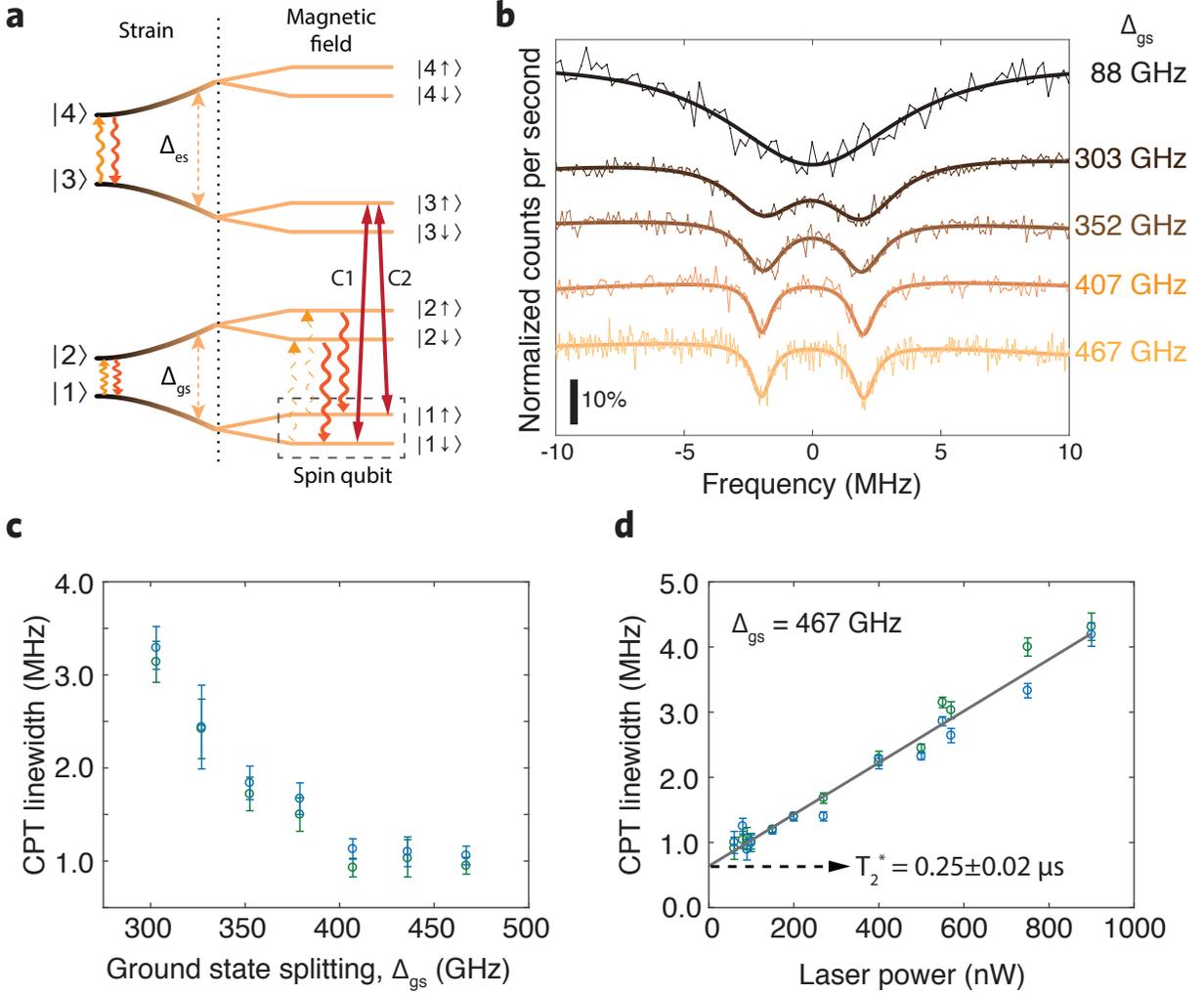


FIG. 3. (a) SiV level structure in the presence of strain and external magnetic field. A spin qubit is defined with levels  $|1\downarrow\rangle$  and  $|1\uparrow\rangle$  on the lower orbital branch of the GS. This qubit can be polarized, and prepared optically using the  $\Lambda$ -scheme provided by transitions C1 and C2. Phonon transitions within ground- and excited-state manifolds are also indicated. The upward phonon transition (phonon absorption process) can be suppressed at high strain, thereby mitigating the effect of phonons on the coherence of the spin qubit. (b) Coherent population trapping (CPT) spectra probing the spin transition at increasing values of the GS orbital splitting  $\Delta_{gs}$  from top to bottom. Bold solid curves are Lorentzian fits. Optical power is adjusted in each measurement to minimize power-broadening. (c) Linewidth of CPT dips (estimated from Lorentzian fits) as a function of GS orbital splitting  $\Delta_{gs}$  indicating improvement in spin coherence with increasing strain. (d) Power dependence of CPT-linewidth at the highest strain condition ( $\Delta_{gs}=467$  GHz). Data points are estimated linewidths from CPT measurements, and the solid curve is a linear fit, which reveals linewidth of  $0.64 \pm 0.06$  MHz corresponding to  $T_2^* = 0.25 \pm 0.02 \mu\text{s}$ .

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