Coherent Control of Spins with Gaussian Acoustics

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Abstract:

Hybrid quantum systems combine the advantages of dissimilar quantum degrees of freedom to solve challenges of communicating between disparate quantum states. Silicon carbide (SiC) is an exemplary platform for hybrid spin-mechanical systems, providing long-lived spin registers within optically-active defects, wafer-scale availability, and low acoustic losses. Past demonstrations of spin-mechanical coupling have used uniaxial strain to drive magnetically forbidden spin transitions. With a Gaussian surface acoustic wave resonator, here we show universal control of divacancy spin qubits using all mechanical degrees of freedom, especially shear. We demonstrate Autler-Townes splitting, coherent mechanically driven Rabi oscillations, and direct optical observation of acoustic paramagnetic resonance. This work expands the versatility of mechanically driven spins and shows promise towards integrating spins with quantum nanomechanical systems.

Main Text:

Hybrid quantum systems\textsuperscript{1} leverage the strengths of various systems, including optical photons for sending quantum states across long distances, spins for information storage, microwave superconducting circuits for computation, while potentially using nanomechanics as an intermediary quantum bus. Coherently exchanging quantum information between optically-active defect spins and mechanical resonators provides a route to hybrid quantum systems\textsuperscript{2} for coupling optical photons to microwave frequency phonons. Optically-active defect spins in silicon carbide (SiC), such as the
neutral divacancy\textsuperscript{3} (VV), have recently demonstrated long-lived spin states\textsuperscript{4,5}, a variety of quantum control protocols\textsuperscript{6}, and single defects for spin-photon interfaces\textsuperscript{7} compatible with quantum entanglement protocols. Importantly, SiC is piezoelectric and supports mature fabrication processes for producing micro-electromechanical systems (MEMS). While progress has been made coupling spins to mechanics in similar defect systems, including the NV center\textsuperscript{8} in diamond with strain tuning\textsuperscript{9,10} and driving\textsuperscript{11,12}, there remain significant challenges to coherently manipulating spins with strain and strongly coupling spins with phonons.

While static strain will generate shifts in ground state energy sublevels, resonant strain can be used to coherently drive magnetically forbidden electron spin transitions. Large in-plane dynamic strains can be generated by surface acoustic wave (SAW) devices, which are well developed for RF filters and offer simple engineering approaches for fabricating low loss resonators. SAW devices have also been proposed as hybrid quantum transducers\textsuperscript{13} and used to demonstrate coupling to superconducting qubits\textsuperscript{14-16} along with optomechanical interactions involving defect excited states\textsuperscript{17}.

Here, we show coherent mechanical driving of VV spin ensembles in 4H-SiC using a patterned Gaussian SAW resonator. We demonstrate Autler-Townes effects, mechanical Rabi oscillations, SAW spatial mode mapping, and acoustically driven $\Delta m_s = \pm 1$ spin transitions recently predicted for point defects\textsuperscript{18}. We also discuss how strain fields interact with defect spins differently from electric fields, and how shear components of the strain tensor play an important role not considered in past experimental spin driving research\textsuperscript{9,10}. This work delivers a more general understanding of the zero-field splitting under strain and access to full mechanical control over the ground state spin, permitting new protocols for quantum sensing with MEMS.

We design and fabricate a SAW resonator in order to create RF mechanical strain in the SiC substrate. Since SiC is weakly piezoelectric, we use a thin, sputtered AlN layer for improved piezoelectric coupling. Standard planar SAW resonator designs span wide apertures, often greater than $100\mu m$, distributing the strain across large crystal areas. Since AlN and 4H-SiC have isotropic in-plane Rayleigh wave velocities\textsuperscript{19} (5790 and 6830 m/s, respectively), we design and fabricate simple Gaussian geometries, inspired by Gaussian optics, to focus strain while also eliminating acoustic diffraction losses. A patterned aluminum interdigitated transducer (IDT) transmits SAWs at an acoustic wavelength ($\lambda$) of 12 $\mu m$, while grooves in AlN form Bragg gratings that act as SAW cavity mirrors (Fig. 1A) to support a resonator frequency $\omega_m/2\pi \approx 560$ MHz. Our Gaussian SAW resonator has a loaded quality factor $\sim16,000$ (Fig. 1B), giving a greater $fQ$ product compared to past work on anisotropic substrates\textsuperscript{20}. To directly visualize the Gaussian mechanical mode, we use stroboscopic scanning X-ray diffraction microscopy\textsuperscript{21} (s-SXDM) for imaging phonons with nanoscale resolution. This technique utilizes synchrotron X-ray pulses to measure local oscillations of lattice curvature from a SAW. Images formed by scanning a nano-focused X-ray beam in real space clearly show Gaussian focusing (Fig. 1C) consistent with the fabricated geometry and approximately nanometer Rayleigh wave displacements. In our experiments Gaussian geometries are necessary to obtain large strains for fast coherent manipulation of electron spin states.

We measure the electron spin ground state sublevels ($S = 1$, and spin projections $m_s = 0, \pm 1$) of VV defects using optically detected magnetic resonance (ODMR) with $\Delta m_s = \pm 1$ transitions magnetically driven by microwave fields. Due to the defect’s intersystem crossing, ODMR probes the spin projections of $|\pm 1\rangle$ versus$|0\rangle$ through changes in photoluminescence (PL). The ground state
spin Hamiltonian takes the form,

$$H/\hbar = \gamma B \cdot S + S \cdot D \cdot S$$

Where $\hbar$ is the Planck constant, $\gamma$ is the electron gyromagnetic ratio ($\approx 2.8$ MHz/G), $B$ is the external magnetic field vector, and $D$ is the zero-field splitting tensor. In the absence of lattice strain the VV spin-spin interaction simplifies to $D_0 S_z^2$ where $D_0 \sim 1.336$ and 1.305 GHz, depicted in Fig. 2A, for c-axis oriented defect configurations $hh$ and $kk$, respectively. The zero-field splitting is sensitive to lattice perturbations such as thermal disorder, an applied electric field, or strain. When the lattice is perturbed by a small strain tensor $\varepsilon_{kl}$, the zero-field splitting tensor is generally modified by $\Delta D_{ij} = G_{ijkl} \varepsilon_{kl}$ where $G_{ijkl}$ is the spin-strain coupling tensor. The symmetry of the spin-strain coupling tensor is determined by the local $C_{3v}$ symmetry of the $hh$ and $kk$ configurations for VV (Fig. S1). Off-diagonal Hamiltonian elements caused by $\Delta D_{ij}$ can be utilized to drive resonant spin transitions with phonons.

In order to confirm coherent mechanical spin driving for magnetically forbidden $\Delta m_s = \pm 2$ transitions, the spins must experience a significant mechanical transition rate ($\Omega_m$) compared to the ensemble inhomogeneous linewidth. For these transitions, PL contrast from ODMR cannot directly measure resonant strain without extra microwaves because PL contrast is insensitive to differences between $|+1\rangle$ and $|-1\rangle$ states. The mechanical drive is instead measured using Autler-Townes (AC Stark) splittings, where in the dressed basis, the new eigenstates are split in energy by $\Omega_m$ that can be observed in the ODMR spectrum. We use a continuous magnetic microwave pump (Rabi frequency $\Omega_{\text{Rabi}} \sim$ MHz) for $|0\rangle$ to $|\pm 1\rangle$ while the SAW is driven at a constant frequency $\omega_m/2\pi$. Dressed state level anti-crossings are most clearly seen when the $|\pm 1\rangle$ spin sublevels are tuned to the SAW resonance frequency. The dressed spin eigenstate energies during 400 mW RF drive on the Gaussian SAW resonator closely match predictions from theory for $\Omega_m/2\pi \approx 4$ MHz (Fig. 2B). Additionally, the Autler-Townes splitting scales linearly with square-root of RF power delivered to the SAW, which is expected as $\Omega_m$ is linearly proportional to strain (Fig. 2C). The resolved Autler-Townes splitting shows that a significant fraction of the entire $kk$ spin ensemble is experiencing a greater SAW drive strength than the ensemble ODMR inhomogeneous linewidth, allowing for measurement of coherent Rabi oscillations.

Next we demonstrate coherent mechanical driving of $kk$ electron spins using the pulse sequence in Fig. 2D to differentiate between populations transferred to $|+1\rangle$ versus $|-1\rangle$ spin states. The spin ensemble inhomogeneous linewidth ($\sim 1$ MHz) and relatively long cavity ring up time ($2Q/\omega_m \approx 16$ $\mu$s) prevent fast mechanical pulsing, so we keep the mechanical drive on continuously. A pair of magnetic field $\pi$ pulses defines the effective mechanical pulse time $\tau$ seen by the spin ensemble (Fig. 2D). We find that three-level system dynamics are necessary to explain the observed mechanical Rabi oscillations shown in Fig. 2E, in particular the ensemble population at $\tau = 0$. The observed Rabi oscillations qualitatively agree with spin simulations predicted using only the ensemble ODMR spectrum, fitted Autler-Townes splitting, and numerical modeling for inhomogeneous mechanical driving in depth from Rayleigh waves (see Supplementary materials). Short Rabi decay times are primarily explained by SAW strain inhomogeneity across the ensemble.

We perform spatial mapping of the Gaussian SAW mode in order to show that $\Delta m_s = \pm 2$ transitions occur due to the mechanical driving and not due to any stray magnetic or electric fields.
We map changes in the Autler-Townes splitting, shown in Fig. 3A, at a constant magnetic field while sweeping laser position. In the resonator’s transverse direction a clear Autler-Townes splitting maximum, and therefore resonant strain amplitude, is observed at the SAW beam waist. $\Omega_m$ as a function of transverse position is well described by a model Gaussian beam waist of the fundamental mode in the current device geometry (FWHM = 3.3λ) and not due to predicted stray electric fields (Fig. S3). Scanning the laser spot longitudinally (Fig. 3B), along the SAW propagation direction, reveals Autler-Townes oscillations at the resonator’s acoustic half wavelength. The mechanical driving oscillations in the $x$ direction are less than 5% peak-to-peak, contrary to expectations from previous theoretical work$^{24}$, so we consider a more complete theoretical understanding of the spin Hamiltonian under strain.

In order to explain our spatial mapping results we use a combination of numerical simulations in conjunction with density functional theory (DFT) calculations of spin-strain interactions. For strains only containing components in the (1110) plane (defined as the $xz$-plane), we find from symmetry arguments that $\Omega_m = \frac{1}{2}(G_{11} - G_{12})\varepsilon_{xx} - 2iG_{14}\varepsilon_{xz}$ for $\Delta m_s = \pm 2$ transitions, where the spin-strain tensor $G$ is written in Voigt notation. The symmetric form and numerical values from DFT can be found in the Supplementary materials. In Fig. 3C we show finite element simulation results for uniaxial strain $\varepsilon_{xx}$ and shear $\varepsilon_{xz}$ for a Rayleigh wave propagating along the $x$ direction. We model the experimental results by converting the strain maps to $\Omega_m$ using $G_{ijkl}$ calculated from DFT, which is then convolved with both an optical point-spread function and estimated spin distribution (see Supplementary materials). In our model, the ensemble spatial averaging causes of $(G_{11} - G_{12})\varepsilon_{xx}$ and $G_{14}\varepsilon_{xz}$ to yield similar $\Omega_m$ magnitudes, respectively. Consequently, in qualitative agreement with theory, we always experimentally measure a non-zero Autler-Townes splitting in Fig. 3B because $\Omega_m$ is proportional to a linear combination of $\varepsilon_{xx}(S_x^2 - S_y^2)$ and $\varepsilon_{xz}(S_xS_y + S_yS_x)$. Furthermore, our model explains the relative $\Omega_m$ amplitudes between $kk$ and $hh$ (4:0:1.1) observed in Fig. 3D, so results for $\Delta m_s = \pm 2$ transitions are well described by the zero-field splitting tensor when the full strain tensor is taken into account. Since PL6 also responds well to SAW driving, mechanical control of SiC spin ensembles can be extended to room temperature$^{25}$.

The point group symmetries of the VV in SiC and the NV center in diamond also allow for non-zero spin-strain coupling terms that lead to mechanically driven $\Delta m_s = \pm 1$ transitions$^{18}$ (derivation in Supplementary materials). In order to probe these transitions, we tune the magnetic field $B_0$ such that the spin $|0\rangle$ to $|--1\rangle$ transition frequency is matched with the SAW resonator (Fig. 4A). It is critical to design an experimental measurement sequence insensitive to stray magnetic fields from current in the IDT, so we use an interlaced RF pump/laser probe sequence while locking in on $B_0$. Spin rotations are primarily driven and detected during the SAW cavity ring down period without RF, although, the spin ensemble will also encounter some residual magnetic resonance when RF is turned back on due to remaining optical-spin polarization. We detect higher PL contrast when the RF drive is matched to our SAW cavity resonance (Fig. 4B), whereas smaller, residual PL contrast is detected when the RF drive is far off SAW resonance. When PL contrast is normalized by ODMR experiments from magnetic driving, the $kk:hh$ mechanical drive ratio is 0.89 ± 0.10, which agrees with our theoretical model and DFT calculations (ratio ~ 1.0) where shear couples stronger to this transition than uniaxial strain. Spatial dependence confirms the PL contrast we measure on resonance matches our Gaussian resonator’s mechanical mode shape (Fig. 4C); conversely,
magnetic field driving from the IDT results in a flat profile. This demonstration of $\Delta m_s = \pm 1$ transitions by phonons enables direct PL contrast (optical detection) of resonant spin-strain coupling for sensing applications.

In summary, we established a Gaussian surface acoustic wave platform for universal spin ground state control. We developed a general theory of anisotropic lattice perturbations, where local defect symmetries are critical to understanding spin-phonon interactions. Particularly, strains and shears have a complex interplay along with crystal orientation dependence, and therefore cannot necessarily be treated as an equivalent electric field vector. Excited state electron orbitals$^{26}$ and spins$^{27}$ could further enhance strain coupling for hybrid quantum systems compared to ground state spins$^{28}$, and strain effects on defect hyperfine couplings have so far remained unexplored. Our demonstrations of coherent mechanical control over divacancy spins in SiC for both $\Delta m_s = \pm 1$ and $\Delta m_s = \pm 2$ transitions provides a basis for both quantum sensing with MEMS as well as strong coupling spins to single phonons for quantum transduction$^1$, spin squeezing$^{29}$, and phonon cooling$^{30}$ applications.

References and Notes
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Author contributions:

Data and materials availability:
All data are available upon request to the corresponding author.

Additional information:
The authors declare no competing interests.

Supplementary materials:
Materials and Methods
Supplementary Text
Figures S1-S7
Tables S1-S2
References (31-47)
Fig. 1. Gaussian SAW resonator. (A) Schematic of the SAW device geometry fabricated on sputtered AlN on a 4H-SiC substrate. Microwaves drives spin transitions mechanically through the SAW resonator (cyan) and magnetically from the backside coplanar waveguide (orange). The top right inset shows a microscope image of the resonator’s acoustic focus ($\lambda = 12 \, \mu m$, $w_0 = 2\lambda$) with red lines illustrating the wave’s out-of-plane displacement. (B) Magnitude (blue) and phase (red) measurements of the 1-port RF reflection of the Gaussian SAW resonator used in spin experiments. (C) Mechanical mode from a similar Gaussian SAW ($\lambda = 19 \, \mu m$, $w_0 = 1.25\lambda$), directly measured with s-SXDM (peak-to-peak $|\partial u_x / \partial x|$ defined as lattice slope) using 4H-SiC [0004] Bragg peak. The mode is marginally distorted due to sample drift during imaging.
Fig. 2. Coherent mechanical driving of $kk$ spin ensembles in silicon carbide. (A) VV ground state illustration with magnetic ($\Omega_{B \pm 1}$) and electromechanical ($\Omega_m$) drives shown. (B) Autler-Townes measurement on a $kk$ ensemble; dressed for $N$ phonons (black) and undressed (white) spin transitions. The mechanically dressed eigenstates and corresponding transitions are split by $\Omega_m$. (C) Mechanical transition rates obtained from Autler-Townes splittings agree with a linear fit to the square-root of drive power. Error bars are 95% confidence intervals from fits. Inset shows an Autler-Townes splitting measurement (black) at $B_0 \approx 100$ G, with Gaussian fits (red) to the VV$^0$ electron spin and weakly coupled nearby nuclear spins. (D) Pulse sequence for mechanically driven Rabi oscillations. (E) Mechanically driven Rabi oscillations at ~ 400, 100, and 25 mW, respectively, and typical error bars are 95% confidence intervals. Simulations are ensemble average predictions with inhomogeneous strain distributions from finite element modeling.
Fig. 3. Spatial mapping mechanical spin driving in a Gaussian SAW resonator. (A) Autler-Townes splitting (left) of $kk |-1\rangle$ sublevel as a function of transverse position at $x = 0$ and mechanical transition rate (right) analyzed from Autler-Townes splittings. The beam waist model is $\exp\left(-\frac{y^2}{w^2}\right)$, using fabrication parameters and a scaled amplitude. (B) Mechanical transition rate (left) as a function of longitudinal position at $y = 0$, plotted with a line through the experimental data. FFT (right) shows a peak and Gaussian fit in red at the expected acoustic periodicity $\lambda/2$ (6 μm). (C) Strain $\varepsilon_{xx}$ and $\varepsilon_{xz}$ of the SAW modeled with COMSOL Multiphysics. (D) Autler-Townes splitting measurements for $kk$, $hh$, and PL6 with $\Omega_m \sim 4.0$, 1.1, 3.4 MHz, respectively, under the same conditions. All error bars are 95% confidence intervals from fitting.
Fig. 4. Mechanically driven $\Delta m_s = \pm 1$ spin transitions. (A) Energy level diagram showing the SAW frequency on resonance with the spin transition between the $|0\rangle$ and $|-1\rangle$ states. (B) (Top) Interlaced pump-probe sequence during magnetic field modulation. (Bottom) PL contrast when RF excitation is on and off cavity resonance ($\omega_m/2\pi = 559.6$ MHz). RF power is 32 mW at sample. (C) Integrated PL contrast from $kk$ resonance (evaluated at $\Delta B_0 = 0$) as a function of the SAW resonator transverse position. RF on-resonance (“$\Omega_m$”) uses the interlaced sequence from (B), whereas off-resonance data (“$\Omega_B$”) uses a continuous, non-interlaced sequence. RF power is 200 mW at sample. All error bars are 95% confidence intervals.