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Optical spin locking of a solid-state qubit
J. H. Bodey, R. Stockill, E. V. Denning, D. A. Gangloff, G. Ethier-Majcher, D. M. Jackson, E. Clarke, M. Hugues, C. Le Gall and M. Atatüre

Quantum control of solid-state spin qubits typically involves pulses in the microwave domain, drawing from the well-developed toolbox of magnetic resonance spectroscopy. Driving a solid-state spin by optical means offers a high-speed alternative, which in the presence of limited spin coherence makes it the preferred approach for high-fidelity quantum control. Bringing the full versatility of magnetic spin resonance to the optical domain requires full phase and amplitude control of the optical fields. Here, we imprint a programmable microwave sequence onto a laser field and perform electron spin resonance in a semiconductor quantum dot via a two-photon Raman process. We show that this approach yields full SU(2) spin control with over 98\% π-rotation fidelity. We then demonstrate its versatility by implementing a particular multi-axis control sequence, known as spin locking. Combined with electron-nuclear Hartmann–Hahn resonances which we also report in this work, this sequence will enable efficient coherent transfer of a quantum state from the electron spin to the mesoscopic nuclear ensemble.

INTRODUCTION
The existence of strong electric dipole transitions enables coherent optical control of matter qubits that is both fast and local. The optical techniques developed to address central spin systems in solids, such as colour centres in diamond and confined spins in semiconductors, typically fall into two categories: the first makes use of ultrashort, broadband, far-detuned pulses to induce quasi-instantaneous qubit rotations in the laboratory frame. Achieving complete quantum control with this technique further requires precisely timed free precession accompanying the optical pulses. The second technique is based on spectrally selective control via a resonantly driven two-photon Raman process and allows full control exclusively through tailoring of the drive field, echoing the versatility of magnetic spin resonance. Despite this attractive flexibility, achieving high-fidelity control using the latter approach has proved challenging due to decoherence induced by the involvement of an excited state for colour centres in diamond and due to nuclei-induced ground-state decoherence for optically active semiconductor quantum dots (QDs). In the case of QDs, the limitation of ground-state coherence can be suppressed by preparing the nuclei in a reduced-fluctuation state. In this Letter, we achieve high-fidelity SU(2) control on a nuclei-prepared QD spin using a tailored waveform imprinted onto an optical field. We then demonstrate the protection of a known quantum-state via an aligned-axis continuous drive, a technique known as spin locking. Finally, by tuning the effective spin-Rabi frequency, we access the electron-nuclear Hartmann–Hahn resonances, which holds promise for proxy control of nuclear states.

RESULTS
Optical electron spin resonance
Our device is an indium gallium arsenide QD, embedded in an n-type Schottky heterostructure and housed in a liquid-Helium cryostat at 4.2 K. Fig. 1a depicts this arrangement. The QD is charged deterministically with a single electron, and a magnetic field of 3.3 T perpendicular to the growth axis creates an \( \omega_b = 24.5 \) GHz Zeeman splitting of the electron spin states which form \( \Lambda \) systems with the two excited trion states. Using an electro-optic modulator (EOM), we access these \( \Lambda \) systems by tailoring a circularly polarised single-frequency laser, of frequency \( \omega_e \) and detuned from the excited states by \( \Delta_L \approx 700 \) GHz. The EOM is driven by an arbitrary waveform generator (AWG) output with amplitude \( V_0 \), frequency \( \omega_{\text{MW}} \) and phase \( \phi_{\text{MW}} \). Operating the EOM in the regime where the microwave field linearly modulates the input optical field, a signal \( V_0 \cos(\omega_{\text{MW}}t + \Delta \phi_{\text{MW}}) \) produces a control field consisting of two frequencies at \( \omega_e \pm \omega_{\text{MW}} \) with a relative phase-offset of \( 2\Delta \phi_{\text{MW}} \). This bichromatic field of amplitude \( \Omega \) drives the two-photon Raman transitions with a Rabi coupling strength \( \Omega = \frac{\Omega^2}{\Delta_L} \) between the electron spin states (see Supplementary Note 2) in the limit \( \left( \Omega/\Delta_L \right)^2 \ll 1 \). The Hamiltonian evolution is given by

\[
\hat{H}_{\text{eff}} = \Omega(\cos(\phi)\hat{S}_z + \sin(\phi)\hat{S}_y) + \delta \hat{S}_z
\]

where \( \hat{S}_i \) are the spin operators in the electron rotating frame, \( \delta \) the two-photon detuning and \( \phi \) the relative phase-offset of the Raman beams. The effect of this Hamiltonian is described geometrically by a precession of the Bloch vector around the Rabi vector \( \{ \Omega \cos(\phi), \Omega \sin(\phi), \delta \} \). We have full SU(2) control over the Rabi vector through the microwave waveform, via the Rabi frequency \( \Omega \propto V_0^2 \), its phase \( \phi = 2\Delta \phi_{\text{MW}} \) and the two-photon detuning \( \delta = \omega_e - 2\omega_{\text{MW}} \). An additional resonant optical field of 100-ns duration performs spin initialisation and readout. Finally, prior to the whole protocol, we implement the recently developed nuclear-spin narrowing scheme, which conveniently requires no additional laser or microwave source, in order to enhance the coherence of the ground state and maximise control fidelity.

Figure 1b shows the evolution of the population of the \( |1\rangle \) state for increasing durations of the Raman drive, taken at different Raman powers. The Raman drive induces coherent Rabi
Coherence of optical rotations

We characterise the coherence of the rotations with the quality factor $Q$, which measures the number of $n$ rotations before the Rabi-oscillation visibility falls below $1/e$ of its initial value. Figure 2a summarises the dependence of the $Q$ factor and decay of the Rabi envelope on the ESR drive strength $\Omega$ and sheds light on three distinct regimes, which are dominated by one of three competing decoherence processes included in the model curve of Fig. 2: (i) inhomogeneous broadening of variance $\sigma = 4.8$ MHz, (ii) electron-mediated nuclear spin–flipping transitions arising from the presence of strain, and (iii) a spin decay proportional to the laser power, which for simplicity we cast as $\Gamma_1 = a [Q]$ with $a = 2.7 \times 10^{-2}$. In the low-power regime, where $\Omega < 18$ MHz, the fidelity is affected by nuclei-induced shot-to-shot detuning errors, which in our model are fixed according to an independent Ramsey measurement. This inhomogeneous broadening induces a non-exponential decay of Rabi oscillation visibility $^7$ (see also Supplementary Note 3). We shield the system from this effect by increasing the Rabi frequency, yielding an increase in $Q$ factor. The intermediate-power regime, where $\Omega = 18 - 80$ MHz, exhibits a dramatic decrease in $Q$ and increase in decay rate. In this regime, the coherent spectrally selective drive induces electron-mediated nuclear spin-flips through a Hartmann–Hahn resonance, $^8$ as we depict in the inset to Fig. 2a. Splitting the dressed electron states $|\uparrow\rangle, |\downarrow\rangle$ by an energy $\hbar \Omega$ causes the dressed electron-nuclear states to become degenerate, removing the energy cost associated to a single nuclear spin–flip $\sim \hbar \Omega \omega_{\text{nuc}}$. The presence of intrinsic strain, which perturbs the nuclear quantisation axis set by the external magnetic field, allows coupling between these now-
degenerate states. The decay of electronic coherence is related to the nuclear spectral density shown in Fig. 2b, which captures the strength of the strain-enabled nuclear transitions over a nuclear ensemble of \( N \approx 74,000 \) nuclei inhomogeneously broadened by variation of the local strain fields across the QD (see Supplementary Note 4). As an intuitive semi-classical picture, one can think of the Knight field—the electron-spin polarisation \( S_J \) felt by the nuclei—acting as an effective radio-frequency field of frequency \( \Omega \) along the external magnetic field. Strain, tilted from this external field, is a perturbation that allows this Knight field to induce single-nucleus transitions between the eigenstates \( \epsilon_m \) and \( \epsilon_{m'} \) to first order (inset of Fig. 2b) provided \( \Omega \) is close to \( \omega_{nc} \) or \( 2\omega_{nc} \). In the high-power regime \( (\Omega > 80 \text{ MHz}) \), we decouple from both inhomogeneous nuclear-spin fluctuations and Hartmann–Hahn transitions, and consequently observe the highest \( Q \) factors \( (Q = 47.6 \pm 1.7 \) over the four highest Rabi frequencies\). Here, the decay envelope is dominated by \( \Gamma_1 \), an optically induced relaxation between the electron states proportional to power, and independent of the single-photon detuning \( \Delta \) (see Supplementary Note 3). The non-resonant and non-radiative nature of this process is consistent with electron-spin relaxation induced by photo-activated charges appearing in our device as Stark shifts of the electronic coherence is related to the nuclear spectral density shown in Fig. 2b, which captures the strength of the strain-enabled nuclear transitions over a nuclear ensemble of \( N \approx 74,000 \) nuclei inhomogeneously broadened by variation of the local strain fields across the QD (see Supplementary Note 4). 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DISCUSSION

The high-fidelity all-optical ESR we report here enables the generation of any quantum superposition spin state on the Bloch sphere using a single waveform-tailored optical pulse. This full SU (2) control further allows the all-optical implementation of spin locking, traditionally an NMR technique, for quantum-state preservation via gapped protection from decoherence-inducing environmental dynamics. In the case of semiconductor QDs, expected from our Fig. 2 model is evidence that spin locking is similarly limited by the photo-activated spin relaxation (Γₘ). The quantum state is thus locked and preserved for a longer time than would be accessible via direct Rabi drive. This confirms our ability to implement all-optically a multi-axis spin-control sequence that can protect the qubit effectively against the intrinsic nuclear hyperfine coupling.

METHODS

Quantum dot device

Our QD device is the one used in ref. 30. Self-assembled InGaAs QDs are grown by molecular beam epitaxy and integrated inside a Schottky diode structure, above a distributed Bragg reflector to maximise photon-outcoupling efficiency. There is a 35-nm tunnel barrier between the n-doped layer and the QDs, and a blocking barrier above the QD layer to prevent charge leakage. The Schottky diode structure is electrically contacted through Ohmic AuGeNi contacts to the n-doped layer and a semitransparent Ti gate (6 nm) is evaporated onto the surface of the sample. The photon collection is enhanced with a superhemispherical cubic zirconia solid immersion lens on the top Schottky contact of the device. We estimate a photon-outcoupling efficiency of 10% at the first lens for QDs with an emission wavelength around 970 nm. A home-built microscope with spectral and polarisation filtering is used for resonance fluorescence, with a QD-to-laser-coupling ratio exceeding 100:1.

Raman laser system

Sidebands are generated from the continuous-wave laser by modulating a fibre-based EOSPACE EOM with a microwave derived from a Tektronix AWG 70002A. The output of the EOM depends on the voltage applied, with maximum and minimum transmissions for applied voltages V_max and V_min, respectively, and the r voltage of the EOM, V_r = |V_max − V_min|. Applying a voltage V = V_min + V_0(t), where V_0(t) is a microwave field of small amplitude compared with V_r, the electric field at the EOM output E_out is described by E_out(t) ∝ V_0(t) × E_in(t). In other words, we work with small amplitude around the minimum intensity transmission of the EOM to imprint the microwave amplitude onto the optical field.

Generation of the microwave signal V_0(t) is depicted in Fig. 5. We produce a digital signal with a sampling rate that is four times the microwave frequency (a factor 2 is obtained by setting the AWG sampling rate at 2ω_mw, and another factor 2 is obtained by combining two independently programmable AWG outputs with a splitter). We thus arrive at a digital signal containing four bits per period, the minimum required to carry phase information to the EOM. To generate the signal shown in Fig. 5, we add the two AWG outputs in quadrature, which we

where the nuclei form the dominant noise source, the same quantum control capability enables us to reveal directly the spectrum of nuclear-spin dynamics. An immediate extension of this work will be to perform spin locking in the spectral window of nuclear-spin resonances, i.e. the Hartmann–Hahn regime, to sculpt collective nuclear-spin states, and also to tailor the electron–nuclear interaction to realise an ancilla qubit or a local quantum register based on the collective states of the nuclear ensemble.

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DISCUSSION

The high-fidelity all-optical ESR we report here enables the generation of any quantum superposition spin state on the Bloch sphere using a single waveform-tailored optical pulse. This full SU (2) control further allows the all-optical implementation of spin locking, traditionally an NMR technique, for quantum-state preservation via gapped protection from decoherence-inducing environmental dynamics. In the case of semiconductor QDs,
Fig. 6 A typical experimental cycle. The nuclear-spin bath is prepared using a Raman cooling technique for 4 μs, before electron spin control is performed. This takes place over a similar timescale, meaning that our duty cycle is close to 50%.

realise after characterisation of the relative delay between the two microwave lines arriving at the splitter. From each output, we generate a square-wave signal at 12.25 GHz. By tuning their relative amplitudes, we construct a sinusoidal signal at 12.25 GHz whose phase ϕ is determined by the relative amplitude A1/2 of channels 1 and 2 according to tan(ϕ) = A1/A2.

Experimental cycle

Nuclear-spin preparation. Figure 6 shows our experimental cycle which involves narrowing the nuclear-spin distribution before a spin-manipulation experiment. Nuclear-spin preparation is done using the scheme detailed in ref. 17, operating in a configuration analogous to Raman cooling in atomic systems. It involves driving the system continuously with the Raman laser, while pumping the |↓⟩ spin state optically. Optimum cooling, assessed using Ramsey interferometry, occurs for a Raman drive at Ω = 22 MHz and a resonant repump of Δ0 = 0.9fO/√2 for an excited-state linewidth Γ0 in agreement with the optimum conditions found in ref. 13. These settings give an order-of-magnitude improvement in our electron spin inhomogeneous dephasing time T2 (Fig. 2a).

Electron spin control. During spin control, we conserve the total Raman pulse area in our sequences by pairing pulses of increasing length with pulses of decreasing length (Fig. 6). This allows us to stabilise the Raman laser power using a PID loop and maintain relative fluctuations below a per cent. We operate with a duty cycle of around 50%, preparing the nuclear-spin bath for a few μs before spending a similar amount of time performing electron spin control. The alteration on μs timescale of coherent manipulation and nuclear-spin preparation is fast compared with the nuclear-spin dynamics15 such that the nuclear-spin distribution is at steady state.

DATA AVAILABILITY

The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

CODE AVAILABILITY

The code used to produce the theoretical findings of this study is available from the corresponding authors upon reasonable request.

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AUTHOR CONTRIBUTIONS
J.H.B., R.S., D.A.G., G.É.-M., D.M.J., C.L.G. and M.A. conceived the experiments. J.H.B., R.S. and C.L.G. acquired and analysed data. E.V.D., C.L.G. and J.H.B. developed the theory and performed simulations. E.C. and M.H. grew the sample. J.H.B., R.S., E.V.D., D.A.G., G.É.-M., D.M.J., C.L.G. and M.A. prepared the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
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