Optical manipulation of the Berry phase in a solid-state spin qubit

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Phase relations between quantum states represent a resource for storing and processing quantum information. Although quantum phases are commonly controlled dynamically by tuning energetic interactions, the use of geometric phases that accumulate during cyclic evolution may offer superior robustness to noise. To date, demonstrations of geometric phase in solid-state systems employ microwave fields that have limited spatial resolution. Here, we demonstrate an all-optical method to accumulate a geometric phase, the Berry phase, in an individual nitrogen-vacancy centre in diamond. Using stimulated Raman adiabatic passage controlled by diffraction-limited laser light, we loop the nitrogen-vacancy centre’s spin around the Bloch sphere to enclose an arbitrary Berry phase. We investigate the limits of this control due to the loss of adiabaticity and decoherence, as well as its robustness to noise introduced into the experimental control parameters. These techniques set the foundation for optical geometric manipulation in photonic networks of solid-state qubits linked and controlled by light.

When a quantum mechanical system evolves slowly along a closed loop in its parameter space, a given eigenstate may acquire a phase consisting of dynamic and geometric contributions. First proposed by S. Pancharatnam in his study of cyclic rotations of light polarization and later generalized by M.V. Berry, this adiabatic geometric phase is determined solely by the geometry of the traversed loop, in contrast to the dynamic phase, which accumulates from the energetics and travel time of the intervening state evolution. Because the Berry phase is proportional to the area enclosed by the path in parameter space, it is intrinsically resilient to noises that deviate the path but conserve the enclosed area. Geometric control thus represents a promising avenue for constructing fault-tolerant quantum logic gates.

Control over geometric phases, occurring both when the cyclic evolution is traversed adiabatically and non-adiabatically, has been demonstrated in a variety of physical platforms, including liquid nuclear magnetic resonance, trapped atoms and recently in avenue for constructing fault-tolerant quantum logic gates. M.V. Berry, this adiabatic geometric phase is determined solely by the geometry of the traversed loop, in contrast to the dynamic phase, which accumulates from the energetics and travel time of the intervening state evolution. Because the Berry phase is proportional to the area enclosed by the path in parameter space, it is intrinsically resilient to noises that deviate the path but conserve the enclosed area. Geometric control thus represents a promising avenue for constructing fault-tolerant quantum logic gates.

Our realization in the solid state offers potentially faster adiabatic control.

The method we employ is based on proposals to accumulate geometric phase via stimulated Raman adiabatic passage (STIRAP) and solid-state systems have used microwaves, which are difficult to localize and thus concede the ability to selectively address nearby qubits without crosstalk. Here, we manipulate the Berry phase in a solid-state qubit using diffraction-limited resonant laser fields. This opens the possibility for independent manipulation of single qubits in photonic networks and spin arrays through geometric principles where fine control over the energetics is not essential. Whereas a similar optical protocol has recently been achieved using trapped calcium ions, our realization in the solid state offers potential integration into photonic platforms and harnesses larger energy separations that enable significantly faster adiabatic control.

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Understanding STIRAP in the NV centre

We exploit a natural A system within the NV centre level structure at cryogenic temperatures (T = 8 K), which is formed by the ground spin states and coupled to the A2 spin–orbit excited state. Applying a ~117 G magnetic field along the NV axis splits and by 655 MHz, tuning this A system into a non-degenerate configuration (Fig. 1c). On-chip microwave
Figure 1 | Driving the $|A_2\rangle$ Λ system. a, Simplified experimental schematic detailing the confocal microscope used to optically manipulate the Berry phase within an NV centre in diamond. A resonant laser, tunable across the NV centre zero-phonon line transitions (≈637 nm), passes through an electro-optic modulator (EOM), where it acquires frequency sidebands and is focused by an objective onto the NV centre circled in red in the photoluminescence (PL) image. b, Photoluminescence excitation (PLE) intensity spectrum displaying the transition to the excited state $|A_2\rangle$ for the ground state spin initialized into $|-1g\rangle$ (red) or $|+1g\rangle$ (blue). c, Λ system within the NV centre level structure consisting of $|-1g\rangle$ and $|+1g\rangle$ coupled to the spin-orbit excited state $|A_2\rangle$ by two optical driving fields $\Omega_1(t)$ and $\Omega_2(t)$, with one-photon detuning $\delta$ and two-photon detuning $\gamma$. d, Evolution of state transfer through STIRAP on the Bloch sphere. This ‘tangerine slice’ trajectory (red to blue gradient) starting and ending at $|a\rangle$ encloses a wedge angle $\Phi$ and results in a Berry phase $\gamma_0$, proportional to $\Phi$, that accumulates on $|a\rangle$. e, Top: Normalized optical Rabi frequencies of driving fields $\Omega_1(t)$ (blue) and $\Omega_2(t)$ (red), enabling the movement of the dark state from the $|-1g\rangle$ pole (t = 0) to the $|+1g\rangle$ pole (t = $\pi/2$) and back (t = $\pi$) during STIRAP. Bottom: Relative phase $\phi(t)$ between the two driving fields. The phase shifts by $\Phi$ at the $|+1g\rangle$ pole (t = $\pi/2$).

control enables rotations between the third ground state $|0g\rangle$ and either $|-1g\rangle$ or $|+1g\rangle$. These microwaves are used only to prepare and read out the spin state (refs 30,31 and Supplementary Section 2.2.2), and not to accumulate geometric phase. Instead, we address the Λ system using a narrow-line tunable 637 nm diode laser (470 THz) fibre-coupled into an electro-optic modulator (EOM). Driving the EOM with a signal generator places frequency harmonics on the laser equivalent to the $|-1g\rangle$/$|+1g\rangle$ splitting (655 MHz) with the relative phase and amplitude of the harmonics controlled on nanosecond timescales. From this, we achieve full Bloch sphere control over the resultant dark state, $|D\rangle$.

In the present experiment, the redshifted first harmonic of the laser is tuned to the $|+1g\rangle$ to $|A_2\rangle$ transition with Rabi coupling strength $\Omega_1(t)$, and the zeroth harmonic is tuned to the $|-1g\rangle$ to $|A_2\rangle$ transition with Rabi coupling strength $\Omega_2(t)$. The two optical fields are deliberately detuned from the one-photon resonance by a redshift of $\Delta = 65 \pm 15$ MHz to limit unintended absorption and maintain fast manipulation during STIRAP (Supplementary Section 2.4.2). We begin by preparing the spin into $|-1g\rangle$. Applying only $\Omega_1$, sets $|-1g\rangle$ as the initial dark state. By adiabatically shifting the relative intensity from $\Omega_1$ to $\Omega_2$, the dark state gradually moves on the surface of the Bloch sphere from $|-1g\rangle$ to the opposite $|+1g\rangle$ pole (Fig. 1d). The precise time evolution of the field amplitudes $\Omega_1(t)$ and $\Omega_2(t)$, displayed in Fig. 1e, is governed by the applied pulse shape and harmonic generation relation in the EOM (Supplementary Section 2.2.1). A phase shift $\Phi$ between the optical fields at the opposite pole ($t = \tau/2$) followed by a reversal of the intensity shift, returns the dark state to the $|-1g\rangle$ pole along a different Bloch sphere longitude and completes a cyclic route in traversal time $\tau$. This ‘tangerine slice’ trajectory with wedge angle $\Phi$ circumscribes a solid angle $2\Phi$, and thus gives rise to an accumulated Berry phase (Fig. 1d,e).

We begin by exploring the mechanisms limiting STIRAP in the NV centre by tomographically reconstructing the path of the spin on the $|-1g\rangle$ to $|+1g\rangle$ Bloch sphere (Fig. 2a). In this particular instance, we demonstrate a trajectory with outbound (red) and inbound (blue) paths separated by $\Phi = 120^\circ$ for an adiabatic cycle time of $\tau = 1,200$ ns and a peak optical Rabi frequency of $\Omega_{\text{R}} = 31 \pm 3$ MHz for the $|-1g\rangle$ transition. From this time-resolved reconstruction, we observe that the length of the dark-state Bloch vector (Fig. 2b) decreases around the equator, revives near the opposite pole, and ultimately returns to the initial pole with 65% of its original magnitude.

This decrease in the state magnitude along the traversal is due to a lag in the adiabatic following of the dark state. Increases in the velocity of the trajectory can cause coupling strengths between the dark and non-dark eigenstates to exceed their energy gap, leading to nonadiabatic evolution. These diabatic transitions into non-dark states reduce the overall magnitude of the state vector, as these states point in different directions with respect to the intended dark state on the $|-1g\rangle$/$|+1g\rangle$ Bloch sphere. Furthermore, as they contain components of the excited state $|A_2\rangle$, absorption can occur and lead to decay either into the intended dark state, causing recovery of the magnitude, or into $|0g\rangle$, causing irreversible, accumulated loss out of the subspace. For our STIRAP pulse shape, the state magnitude decreases where the velocity of the trajectory is greatest (near the equator), and these dips partially recover as the velocity of the path slows near the $|+1g\rangle$ pole. A four-state master equation model (see Methods and Supplementary Section 1) reproduces the salient features of the trajectory in Fig. 2b. Finally, Fig. 2c plots the inbound trajectories for paths enclosing $\Phi = 0^\circ$ to $330^\circ$ in $30^\circ$ increments, demonstrating the ability to enclose any wedge angle $\Phi$.

The photoluminescence during the STIRAP interaction further qualifies the adiabaticity of the path. Non-adiabatic evolution permits excitation to $|A_2\rangle$ and emitted photons, while adiabatic evolution remains dark. The time-resolved photoluminescence during STIRAP (Fig. 2d, blue) indicates that the interaction is dark when compared with a non-adiabatic interaction that optically pumps the spin from $|-1g\rangle$ to $|+1g\rangle$ and then pumps the spin back to $|-1g\rangle$ midway through the interaction (Fig. 2d, red), a form of coherent population trapping (CPT). Indeed, the average number of photons emitted during the STIRAP interaction is nine times fewer than the number of photons emitted at the beginning of the CPT interaction when the population is maximally inverted. Notably, we observe that additional photon emission (Fig. 2d) coincides with a reduction in the dark-state vector (Fig. 2b), both consequences of the loss of adiabatic following.

Optical accumulation of the Berry phase

With the ability to enclose loops of arbitrary wedge angle $\Phi$ on the $|-1g\rangle$/$|+1g\rangle$ Bloch sphere, we extend this STIRAP technique to...
observe the Berry phase $\gamma_b$, accumulated on $|{-1}_y\rangle$ after a cycle has completed. Using the third ground spin state $|0_y\rangle$ as a phase reference, we place the spin into a fixed $|0_y\rangle/|{-1}_y\rangle$ superposition and then use STIRAP to enclose a given $\Phi$ within the $|{-1}_y\rangle/|{+1}_y\rangle$ subspace. State tomography determines the phase that accumulates on $|{-1}_y\rangle$ relative to $|0_y\rangle$ for the final superposition state (Fig. 3a). This accumulated phase can contain contributions from both a dynamic phase offset $\eta$, independent of the wedge
The dark state magnitude of 65% (in Fig. 2), as non-adiabatic transitions with a dynamic phase offset of 59°. The amplitude of the oscillations expectedly shows that the Berry phase indeed matches the acquired phase. Fitting these projections collectively, we determine that the dark state does not couple to the light fields and experiences no Stark shift. Precisely setting δ = 0 through spectroscopic measurements is difficult (an estimated detuning of Δ = 250 kHz is evident in Fig. 3b to show the dynamic phase contribution). However, by measuring η(τ) as a function of the sideband separation, we extract the optical Stark frequency shift, Σ/δ = (1/360°)(dη(τ)/dδ), and isolate where Σ = 0 for δ = 0. In Supplementary Section 2.3.3 we present Berry phase traces calibrated to two-photon resonance (Δ < 3 kHz) that display virtually no dynamic phase accumulation. Analysing similar traces in Fig. 4a and its inset, we find that Σ scales linearly in δ. Perturbation theory in small δ shows that this ratio is independent of Ω, with an expected relation of Σ = 0.556 for our pulse shape, matching the experimental result well (Fig. 4a, inset). Unlike the dynamic phase, which requires fine control of δ and τ, the Berry phase has no dependence on these parameters as long as the STIRAP interaction remains adiabatic.

Limits and robustness of the Berry phase

To facilitate comparative measurements of the visibility and isolate the effects of control noise on the Berry phase, we implement a Hahn echo sequence20,43 (Supplementary Section 2.4.1), where a microwave π pulse separates two time-reversed STIRAP interactions. This sequence doubles the Berry phase, but cancels the dynamic phase accumulation of a single interaction. Examining the visibility of the echoed Berry phase as a function of the traversal time τ and the Rabi frequency Ω, we find a sharp decrease in the visibility where adiabaticity is completely lost for short τ. Similarly, the gradual reduction in the visibility for longer τ is due to non-ideal processes that increase the probability of cycling through |A2⟩ for a given iteration. As we increase Ω from 64 MHz, we achieve visibilities as high as 51% and adiabatic interaction times as short as τ ≈ 250 ns. Faster adiabatic evolution is enabled by increasing Ω, as the energy gap between dark and non-dark states expands. Modelling these trends using our four-state master equation approach, including decoherence and unintended excitation by far-detuned harmonics, is presented in Supplementary Section 1.4. Our fastest geometric control with STIRAP represents a 400-fold speed-up over the previous atomic demonstration27, and is enabled by the larger Zeeman splitting between the two lower levels of the NV centre Λ system.

Furthermore, as the Berry phase arises from global geometric properties of the state evolution, it offers robustness to noises that act locally on the trajectory. To investigate, we introduce simulated noise onto the parameters controlling the polar θ and azimuthal φ angles for our loops. The two types of noise, δθ(t) and δφ(t) acting perpendicular to the ideal path and δθ(t) acting parallel to the ideal path, physically correspond to fluctuations in the relative phase and amplitude, respectively, of the two STIRAP laser fields (Fig. 5a). We measure the standard deviation σθ,φ of the distribution of Berry phases realized from 250 unique instances of noisy paths.

The noises conform to an Ornstein–Uhlenbeck process with a Lorentzian frequency bandwidth of Δν = 3 MHz and a Gaussian distribution of amplitudes with standard deviation s(=θ,φ). Figure 5b plots the distributions (including broadening by photon collection statistics) arising from a noise amplitude of sθ,φ = 8° for
ideal loops enclosing four disparate Berry phase angles. We find these distributions remain constant regardless of the intended Berry phase. This feature results from the symmetric, path-length-preserving nature of our class of trajectories (‘tangerine slice’), as the sensitivity to fluctuations does not depend on the given \( \Phi \). This property is in contrast to other classes of trajectories (paths of variable radius) typically accessed by rotating field approaches\(^5,10,14\), where larger Berry phases are more susceptible to noise.

In Fig. 5c, we examine the impact of increasing the amplitude of the two different noise types. Consistent with the expectation that parallel noise does not change the enclosed solid angle, the measured Berry phases remain minimally dephased for increased \( \delta \theta \) noise. In the case of perpendicular noise, \( \delta \phi \), which modifies the enclosed solid angle, we see an enhanced effect on the angular distribution of the Berry phases. In both cases, larger noise amplitudes reduce the visibility as fewer adiabatic loops are preserved. Assuming the dark state adiabatically follows the noisy path\(^3\), we derive an analytic relationship between the variance in the Berry phase and the noise amplitude \( \sigma_{B} \) for our specific trajectory (Supplementary Section 2.4.5):

\[
\sigma_{B}^2 = \frac{\pi}{2} \frac{1 - e^{-2\Delta t \nu}}{(1 + (\Delta t \nu)^2)^2} \frac{\pi \Delta t \nu}{1 + (\Delta t \nu)^2}
\]

This variance does not depend on wedge angle \( \Phi \), but only on the product \( \Delta t \nu \), a measure of the number of noise oscillations per cycle. In Supplementary Section 2.4.4 we describe the estimation of the intrinsic \( \sigma_{\delta B} \) from the shot-noise-broadened standard deviations \( \hat{\sigma}_{B,echo} \). From this, we confirm in Fig. 5d that \( \sigma_{\delta B} \) is strongly robust to \( \delta \theta \) noise, while its dependence on \( \delta \phi \) noise matches well the expected result \( \sigma_{\delta \theta} = 0.64 \delta \phi \) from equation (2) (solid line in Fig. 5d), using the experimental parameters \( \Delta \nu = 3 \, \text{MHz} \) and \( \tau = 1,200 \, \text{ns} \). In contrast to dynamic phase, the adiabatic geometric phase becomes increasingly robust to noise as the traversal time increases, as seen in \( \sigma_{\delta \theta} \rightarrow (\pi \nu)^2 / (2 \Delta t \nu) \) for \( \Delta t \nu \gg 1 \). Figure 5e displays the estimated \( \sigma_{\delta B} \) as a function of traversal time \( \tau \) for measurements at \( \Delta \nu = 3 \, \text{MHz} \) and a constant noise amplitude \( \delta \phi = 14^\circ \), demonstrating the predicted \( \sigma_{\delta B} \sim \tau^{-1/2} \) scaling that is the hallmark of noise resiliency for geometric phases.

**Conclusions and discussion**

We have demonstrated an all-optical approach to accumulate Berry phase in a solid-state system that enables independent, geometric manipulation of individual qubits with diffraction-limited spatial resolution. Using the \( |\Lambda_2\rangle \Lambda \) system of the NV centre in diamond, we control the adiabatic passage of a dark state, understand the mechanisms that limit the successful enclosure of the Berry phase, and characterize the nature of its robustness to noise. Due to imperfect initialization and loss mechanisms, the experimental Berry phase visibilities peak at 51%, corresponding to an estimated peak state fidelity of 73%. This fidelity is lower than previous geometric phase demonstrations in atomic systems\(^9,17\) and in the solid state using microwaves\(^10-14\). However, we estimate this fidelity could be improved substantially by limiting the effect of far-detuned harmonics, either through increased ground state Zeeman splitting or extinction of unwanted optical drives via cavity rejection and polarization selectivity (Supplementary Sections 1.4 and 1.5). Extensions to this technique could be realized by harnessing other solid-state \( \Lambda \) systems, such as in the silicon–vacancy in diamond\(^35,36\) with its strong zero-phonon line emission, which is important for photonic applications\(^37\). Alternatively, adding another optical field to actively control the third ground-state level in a solid-state tripod system provides an avenue for all-optical universal geometric single qubit gates\(^6,17-19\). The prevalence of \( \Lambda \) and tripod energy structures...
makes these techniques extendable to a variety of solid-state qubits, including colour centres,35,38–40, transition-metal41 or rare-earth ions42,43, and quantum dots44 existing in materials45 that are promising for photonic technologies.

Methods

Methods and any associated references are available in the online version of the paper.

Received 16 June 2015; accepted 14 December 2015; published online 15 February 2016

References

1. Pancharatnam, S. Generalized theory of interference, and its applications. Proc. Indian Acad. Sci. A 44, 247–262 (1956).

2. Berry, M. V. Quantal phase factors accompanying adiabatic changes. Proc. R. Soc. Lond. A 392, 45–57 (1984).

3. De Chiara, G. & Palma, G. M. Berry phase for a spin 1/2 particle in a classical fluctuating field. Phys. Rev. Lett. 91, 090404 (2003).

4. Berger, S. et al. Exploring the effect of noise on the Berry phase. Phys. Rev. A 87, 060303 (2013).

5. Zanardi, P. & Rasetti, M. Holonomic quantum computation. Phys. Lett. A 264, 94–99 (1999).

6. Duan, L. M., Cirac, J. I. & Zoller, P. Geometric manipulation of trapped ions for photonic technologies. Phys. Rev. Lett. 114, 053603 (2015).

7. Toth, M. R. & Sucher, M. Holonomic quantum computation in a quantum dot. Phys. Rev. A 89, 062302 (2014).

8. Jones, J., Vedral, V., Ekert, A. & Castagnoli, G. Geometric quantum computation using nuclear magnetic resonance. Nature 403, 869–871 (2000).

9. Leibfried, D. et al. Experimental demonstration of a robust, high-fidelity geometric two-ion qubit phase gate. Nature 422, 412–415 (2003).

10. Leek, P. J. et al. Observation of Berry’s phase in a solid-state qubit. Science 318, 1889–1892 (2007).

11. Abdumalikov, A. A. Jr et al. Experimental realization of non-Abelian non-adiabatic geometric gates. Phys. Rev. Lett. 115, 080503 (2015).

12. Zu, C. et al. Experimental realization of universal geometric quantum gates with solid-state spins. Nature 514, 72–75 (2015).

13. Arroyo-Camejo, S., Lazarev, A., Hell, S. W. & Balasubramanian, G. Room temperature high-fidelity holonomic single-qubit gate on a solid-state spin. Nature Commun. 5, 4870 (2014).

14. Zhang, K., Nusran, N. M., Slezak, B. R. & Dutt, M. V. G. Measurement of the Berry phase in a single solid-state spin qubit. Preprint at http://arxiv.org/abs/1410.2791 (2014).

15. Lončar, M. & Faraon, A. Quantum photonic networks in diamond. MRS Bull. 38, 144–148 (2013).

16. Toyli, D. M., Weis, C. D., Fuchs, G. D., Schenkel, T. & Awschalom, D. D. Chip-scale nanofabrication of single spins and spin arrays in diamond. Nano Lett. 10, 3168–3172 (2010).

17. Toyoda, K., Uchida, K., Noguchi, A., Haze, S. & Urabe, S. Realization of holonomic single-qubit operations. Phys. Rev. A 87, 052306 (2013).

18. Kis, Z. & Renzoni, F. Qubit rotation by stimulated Raman adiabatic passage. Phys. Rev. A 65, 032318 (2002).

19. Möller, D., Madsen, L. B. & Mølmer, K. Geometric phase gates based on stimulated Raman adiabatic passage in tripod systems. Phys. Rev. A 75, 062302 (2007).

20. Gaubatz, U., Rudecki, P., Schiemann, S. & Bergmann, K. Population transfer between molecular vibrational levels by stimulated Raman scattering with partially overlapping laser fields. A new concept and experimental results. J. Chem. Phys. 92, 5363–5376 (1990).

21. Bergmann, K., Theuer, H. & Shore, B. W. Coherent population transfer among quantum states of atoms and molecules. Rev. Mod. Phys. 70, 1003–1025 (1998).

22. Goto, H. & Ichimura, K. Population transfer via stimulated Raman adiabatic passage in a solid. Phys. Rev. A 74, 053410 (2006).

23. Golter, D. A. & Wang, H. Optically driven Rabi oscillations and adiabatic passage of single electron spins in diamond. Phys. Rev. Lett. 112, 116403 (2014).

24. Doherty, M. W. et al. The nitrogen-vacancy colour centre in diamond. Phys. Rep. 528, 1–45 (2013).

25. Batálov, A. et al. Low temperature studies of the excited-state structure of negatively charged nitrogen-vacancy color centers in diamond. Phys. Rev. Lett. 102, 195506 (2009).

26. Gao, W. B., Imanoglu, A., Bernien, H. & Hanson, R. Coherent manipulation, measurement and entanglement of individual solid-state spins using optical fields. Nature Photon. 9, 363–373 (2015).

27. Togan, E. et al. Quantum entanglement between an optical photon and a solid-state spin qubit. Nature 466, 730–734 (2010).

28. Kosaka, H. & Niiura, N. Entangled absorption of a single photon with a single spin in diamond. Phys. Rev. Lett. 114, 053603 (2015).

29. Buckley, B. R., Fuchs, G. D., Bassett, L. C. & Awschalom, D. D. Spin-light coherence for single-spin measurement and control in diamond. Science 330, 1212–1215 (2010).

30. Yale, C. G. et al. All-optical control of a solid-state spin using coherent dark states. Proc. Natl Acad. Sci. USA 110, 7595–7600 (2013).

31. Bassett, L. C. et al. Ultrafast optical control of orbital and spin dynamics in a solid-state defect. Science 345, 1333–1337 (2014).

32. Bernien, H. et al. Heralded entanglement between solid-state qubits separated by three metres. Nature 497, 86–90 (2013).

33. Pfaff, W. et al. Unconditional quantum teleportation between distant solid-state quantum bits. Science 345, 532–535 (2014).

34. Santori, C. et al. Coherent population trapping of single spins in diamond under optical excitation. Phys. Rev. Lett. 97, 247401 (2006).

35. Pingault, B. et al. All-optical formation of coherent dark states of silicon-vacancy spins in diamond. Phys. Rev. Lett. 113, 263601 (2014).

36. Rogers, L. J. et al. All-optical initialization, readout, and coherent preparation of single silicon-vacancy spins in diamond. Phys. Rev. Lett. 113, 263602 (2014).

37. Riedrich-Möller, J. et al. Deterministic coupling of a single silicon-vacancy color center to a photonic crystal cavity in diamond. Nano Lett. 14, 5281–5287 (2014).

38. Christle, D. J. et al. Isolated electron spins in silicon carbide with millisecond coherence times. Nature Mater. 14, 160–163 (2015).

39. Widmann, M. et al. Coherent control of single spins in silicon carbide at room temperature. Nature Mater. 14, 164–168 (2015).

40. Jungwirth, N. R. et al. A single-molecule approach to ZnO defect studies: single photons and single defects. J. Appl. Phys. 116, 043509 (2014).

41. Kolesov, R. Coherent population trapping in a crystalline solid at room temperature. Phys. Rev. A 72, 051801 (2005).

42. Xia, K. et al. All-optical preparation of coherent dark states of a single rare earth ion spin in a crystal. Phys. Rev. Lett. 115, 093602 (2015).

43. Hansom, J. et al. Environment-assisted quantum control of a solid-state spin via coherent dark states. Nature Phys. 10, 725–730 (2014).

44. López, C. Materials aspects of photonic crystals. Adv. Mater. 15, 1679–1704 (2003).

Acknowledgements

The authors thank C.P. Anderson, B.B. Buckley, D.J. Christle and C.F. de las Casas for discussions and H.L. Bretscher for experimental assistance. This work was supported by the Air Force Office of Scientific Research (FA9550-14-1-0231 and FA9550-15-1-0029), the National Science Foundation (NSF-DMR-1306300) and the German Research Foundation (SFB 767).

Author contributions

C.G.Y., F.J.H. and B.B.Z. performed the experiments. A.A. and G.B. developed the theoretical simulations. All authors contributed to the data analysis and writing of the paper.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to D.D.A.

Competing financial interests

The authors declare no competing financial interests.
Methods
Experimental set-up. The experiments in this work used an electronic-grade diamond substrate purchased from Element Six, measuring $2 \times 2 \times 0.5$ mm. All NV centres in the sample were formed naturally during the growth process. Te:Au (10 nm Ti, 100 nm Au) short-terminated waveguides were lithographically patterned on the surface to provide on-chip microwave control of the NV centres. The sample was thermally sunk inside a liquid helium flow cryostat held at 8 K. The short-terminated waveguide was wire-bonded to a microwave line within the cryostat and connected to the signal generators via a coaxial port. The cryostat served as the sample chamber for a confocal microscopy set-up designed to study individual NV centres. The NV centre studied had a natural optical linewidth of ∼100 MHz with an orbital strain splitting around 7.4 GHz. An applied external magnetic field of 11.7 G split $|\pm_1\rangle$ and $|\pm_2\rangle$ by 655 MHz, and the combination of the natural strain and applied magnetic field split the $|A\rangle$ and $|A\rangle$ excited states by $\sim$2.9 GHz. Through time-resolved measurements of spin populations as a function of the optical interaction time, we determined the branching ratios of $|A\rangle$ to be 11.39 and 50% for decay into the $|0\rangle$, $|1\rangle$ and $|+1\rangle$, respectively (Supplementary Section 2.1.2).

The associated radiative lifetimes for these branching ratios were used in the model described in the following section. The optical Rabi powers used in the experiment (tens of MHz) were larger than the hyperfine coupling to the intrinsic nitrogen nuclear spin (∼2 MHz in the ground state) and thus any given iteration of STIRAP would manipulate the NV centre spin, regardless of the nuclear state. The confocal microscopy set-up consisted of a 532 nm laser to re-ionize the NV$^+$ charge state and initialize to $|3\rangle$, a tunable 637 nm laser tuned to the $|1\rangle$ transition for readout of the spin state and a second tunable 637 nm laser fibre-coupled to an EOM tuned to the $|A\rangle$ transition for the STIRAP interaction. The EOM was driven by a signal generator tuned to 655 MHz, the splitting of $|\pm_1\rangle$ and $|\pm_2\rangle$, creating sidebands on the laser to drive the $A$ transitions. All lasers were controlled using acousto-optic modulators for nanosecond timescale pulsing. All three lasers passed through individual polarization optics and were combined using beamsplitters and dichroic mirrors. The combined beam was eventually focused onto the sample using a 0.85 NA ×100 objective that was aberration-corrected for the cryostat window. Note that a peak Rabi frequency of 31 MHz from the interaction laser corresponds to a laser power of ∼12 µW at the back of the objective. The redshifted phonon sideband of the NV centre’s photoluminescence was spectrally filtered through a series of dichroic mirrors and bandpass filters and then counted in a silicon avalanche photodiode. Those counts were binned via a series of logic switches and then summed by either a time-correlated counting card (Fig. 2d) or a data acquisition card (rest of Fig. 2 and Figs 3–5).

In addition to the 655 MHz applied to the EOM, additional microwave frequencies were needed for characterization. For the Berry phase measurements (Figs 3–5), a second signal generator provided on-chip microwaves tuned to 2.550 GHz, the splitting of the ground state $|0\rangle$ and $|\pm_2\rangle$ levels. However, for the STIRAP path evaluation measurements (Fig. 2), three colours of microwave were required. In this case, the second signal generator provided microwaves tuned to 3.205 GHz, the splitting of $|0\rangle$ and $|\pm_1\rangle$, while a frequency mixer combined the two initial frequencies to provide the third frequency, 2.550 GHz, the $|0\rangle$/$|\pm_1\rangle$ splitting. To phase-control the microwaves, we used the internal IQ modulation functionality of both signal generators. All timing and pulse sequences (Supplementary Sections 2.2.2, 2.3.1 and 2.4.1) were controlled with a 1 GS s$^{-1}$ arbitrary waveform generator.

Theoretical methods. The rotating frame Hamiltonian describing the A system and optical fields within the NV centre level structure is given by

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 2\Delta & \Omega_\delta(t) e^{i\delta(t)} \\ 0 & \Omega_\delta(t) & \Omega_\delta(t) e^{i\delta(t)} & 2\Delta \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

where the matrix representation is given in the basis $|0\rangle, |\pm_1\rangle, |A\rangle, |A\rangle$. The master equation in Lindblad form is given by

$$\rho = -\frac{\gamma}{\hbar} [H, \rho] + \sum_k \left( I_k \rho I_k^\dagger - \frac{1}{2} I_k^\dagger I_k \rho - \frac{1}{2} \rho I_k^\dagger I_k \right)$$

where $I_k$ denote the Lindblad operators describing dissipative processes. These include experimentally estimated relaxation times from $|A\rangle$ to $|\pm_1\rangle$ of ∼31 ns, $|A\rangle$ to $|\pm_2\rangle$ of ∼24 ns, $|A\rangle$ to the reference state $|0\rangle$ of ∼104 ns, an orbital dephasing rate of 7 ns (ref. 31) and an assumed phenomenological spin dephasing rate of 4 μs.

The optical fields in the simulation are described by $\delta(t)$ where the matrix representation is given in the basis $|0\rangle, |\pm_1\rangle, |A\rangle, |A\rangle$. The master equation in Lindblad form is given by

$$\rho = -\frac{\gamma}{\hbar} [H, \rho] + \sum_k \left( I_k \rho I_k^\dagger - \frac{1}{2} I_k^\dagger I_k \rho - \frac{1}{2} \rho I_k^\dagger I_k \right)$$

where $I_k$ denote the Lindblad operators describing dissipative processes. These include experimentally estimated relaxation times from $|A\rangle$ to $|\pm_1\rangle$ of ∼31 ns, $|A\rangle$ to $|\pm_2\rangle$ of ∼24 ns, $|A\rangle$ to the reference state $|0\rangle$ of ∼104 ns, an orbital dephasing rate of 7 ns (ref. 31) and an assumed phenomenological spin dephasing rate of 4 μs.

The optical fields in the simulation are described by $\Omega_\delta(t)$ where the vector $B = \frac{\Omega_\delta(t)}{\Omega_\delta(t)} \cdot \mathbf{\Phi}$ describes the surface of the Bloch sphere. Substituting our dark state, the Berry phase simplifies to

$$\gamma_B = \frac{1}{2} \delta(t) \left( |0\rangle \langle 0| + 1\rangle \langle 1| \right)$$

where the integral is taken over the closed loop on the Bloch sphere.

References
45. Robledo, L. et al. High-fidelity projective read-out of a solid-state spin quantum register. *Nature* 477, 574–578 (2011).