



All-optical control of a single electron spin in diamond

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Precise coherent control of the individual electronic spins associated with atomlike impurities in the solid state is essential for applications in quantum information processing and quantum metrology. We demonstrate all-optical initialization, fast coherent manipulation, and readout of the electronic spin of the negatively charged nitrogen-vacancy (NV⁻) center in diamond at $T \sim 7$ K. We then present the observation of a novel double-dark resonance in the spectroscopy of an individual NV center. These techniques open the door for new applications ranging from robust manipulation of spin states using geometric quantum gates to quantum sensing and information processing.

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The negatively charged nitrogen-vacancy (NV⁻) center in diamond is an atomlike impurity in the solid state that combines coherent optical transitions with a triplet ground state that supports long-lived spin coherences. A number of recent experiments and novel applications have been enabled by the use of a combination of visible frequency lasers to address the electronic orbital states and microwave manipulation to address the spin degree of freedom [1–6]. While techniques for microwave spin manipulation of NV centers are well established, a number of new potential applications could be enabled by controlling the ground state spin sublevels using optical Raman transitions as is done with isolated neutral atoms and ions. For example, the use of multiple Raman transitions between four-level tripod systems was proposed for realization of robust geometrical quantum gates [7,8]. In addition, optical manipulation offers possibilities for improving NV-based metrology applications, for example, by providing access to forbidden transitions between spin sublevels that are more sensitive to magnetic fields [9,10]. Moreover, all-optical manipulation techniques are important for the development of integrated nanophotonic systems for diamond-based scalable quantum optical devices and quantum networks [11–13]. In these devices, microwave structures on the diamond substrate are often incompatible with the fabrication process for the nanophotonic devices, while the use of external microwave sources defeat the scalability of on-chip photonic devices. The complex level structure and selection rules of the NV center's optical transitions offer a rich and flexible set of possibilities for coherent all-optical control of all three spin sublevels. Past experiments have demonstrated optical spin manipulation under large applied strain, electric, or magnetic fields [14–16]. Recently, two-photon Rabi oscillations and stimulated Raman adiabatic passage on microsecond time scales have been demonstrated at ambient fields [17].

In this Rapid Communication, we demonstrate complete all-optical coherent manipulation of the NV spin states. Importantly, initialization and readout of the spin states are also performed all-optically, providing a full set of experimental techniques that eliminates the need for microwave addressing. In addition, we report observations of nearly degenerate dark states associated with a pair of orthogonally polarized dark resonances in a single quantum emitter [18–22]. Such

individually addressable dark states involving all three sublevels of the ground state manifold open up possibilities for robust control of the entire spin-triplet manifold using geometrical Berry phases [7,8].

Our experiments make use of a macroscopic hemisphere of (100) oriented single crystal chemical vapor deposition diamond kept at ~ 7 K in a helium flow cryostat [23]. The hemisphere acts as a solid immersion lens that increases the efficiency of laser excitation and photon collection. An off-resonant 532 nm laser was used for spin and charge state initialization. In addition, three external cavity diode lasers at 637 nm were used for resonant addressing of various optical transitions, which allows for state-selective coherent manipulations. All lasers are pulsed using acousto-optic modulators, with additional electro-optical modulators (EOM) for generating short pulses with fast rise and fall times on the 637 nm lasers as needed. For initial characterization of the system and during spectroscopy of the tripod system, a permanent magnet outside the cryostat was used to generate a Zeeman splitting of the $|\pm 1\rangle$ states. A 15 μm wire under the bottom face of the sample was used to apply microwave pulses, again only for initial characterization.

Figure 1 presents spectroscopy of the NV center that was used in our experiments. The possible optical transitions between the ground and excited states of the NV center are shown in the level diagram in Fig. 1(a), including spin-preserving transitions (solid arrows) and non-spin-preserving transitions (dashed arrows). All of these transitions can be identified in Fig. 1(b), where a cw microwave drive was applied between the $|0\rangle$ and $|\pm 1\rangle$ ground states and a 637 nm laser was scanned across the zero-phonon transitions while photons were collected on the phonon sideband (PSB). To further confirm the identification of the observed transitions, we plot in Fig. 1(c) the frequencies of the peaks found in Fig. 1(b) (black dots) on top of the strain dependence of all the optical transitions. The latter was obtained by diagonalizing the full Hamiltonian for the optically excited states of the NV center [24,25]. In the following experiments, we make extensive use of the transitions involving the A_2 excited state [(black arrows in Figs. 1(a) and 1(b)], while spin initialization and readout involve in addition the $|0\rangle \rightarrow |E_x\rangle$ transition.

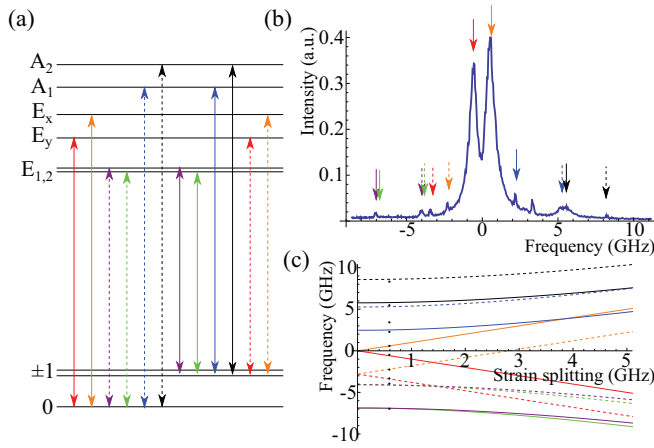


FIG. 1. (Color online) Optical transitions of the NV center at 7 K. (a) All possible transitions between the ground and excited states, with direct transitions indicated with solid lines, and spin nonconserving cross transitions indicated with dashed lines. (b) Photoluminescence spectrum taken with cw microwave excitation. (c) Frequencies of all possible transitions shown in (a) as a function of intrinsic crystal strain in units of the splitting between $|E_x\rangle$ and $|E_y\rangle$. The frequencies of the peaks in (b) are matched to a particular strain value (black dots). The extra unidentified peak may be due to a two-photon transition from the $|0\rangle$ state to the $|E_x\rangle$ state through the absorption of an optical photon and emission of a microwave photon.

To demonstrate all-optical control we first initialize the NV center into an arbitrary ground state spin in zero magnetic field using optical pumping. For example, to prepare the $|-1\rangle$ state, we first apply a laser pulse for $20 \mu\text{s}$ on the $|0\rangle \rightarrow |E_x\rangle$ transition. This optically pumps the spin states into $|\pm 1\rangle$ through non-spin-preserving cross transitions. We then apply a σ_- polarized laser to selectively excite the $|+1\rangle \rightarrow |A_2\rangle$ transition for 400 ns. Using this method, we find that a spin polarization of $> 80\%$ in the $|-1\rangle$ state can be achieved. Similarly, the state can be pre-prepared in the $|+1\rangle$ state by changing the polarization of the laser to σ_+ in the second step. The effectiveness of this method is limited by off-resonant excitation of the $|\pm 1\rangle$ during the first pumping step, decay back into the $|0\rangle$ state during the second pumping step, and imperfect state selection during the second step. In addition, the long optical pumping step on the $|0\rangle \rightarrow |E_x\rangle$ transition can result in ionization of the NV center. We note that the efficiency and fidelity of the optical spin initialization process can be improved by optimizing the length and power of the pumping steps or electrically tuning the NV center to give a more favorable combination of cross transition rates [26].

In the case of zero Zeeman splitting, it is generally difficult to prepare the NV center in a well-defined superposition of the $|\pm 1\rangle$ states using microwave fields, whose polarizations are difficult to control [27]. Using optical initialization, however, an arbitrary superposition of the $|\pm 1\rangle$ states can be prepared by simply changing the polarization of the laser addressing the $|A_2\rangle$ state. This optical pumping scheme can perform well as long as the Zeeman splitting is smaller than the lifetime of the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition. At higher fields where the $|\pm 1\rangle$ states become optically resolvable, preparation in either of the individual $|+1\rangle$ or $|-1\rangle$ states would be unaffected, while

preparation in an arbitrary superposition is possible with two phase-locked lasers at different frequencies.

The second element in all-optical control is the ability to read out any arbitrary spin state. The population of the $|0\rangle$ state can be read out using off-resonant excitation, while resonant single-shot readout of the $|0\rangle$ state has also been demonstrated [26]. To measure the population in some chosen superposition of the $|\pm 1\rangle$ states, one can first transfer it to the $|0\rangle$ state using, e.g., microwave manipulation. A similar technique could be adopted for all-optical readout by coherently transferring $|\pm 1\rangle$ to $|0\rangle$ using Raman transitions (see discussion below). Here, however, we use a simpler method of directly measuring the $|\pm 1\rangle$ population. Since, unlike the $|m_s = 0\rangle$ states, the transitions involving the $|m_s = \pm 1\rangle$ states are not cycling, a long readout pulse will quickly optically pump the population into the orthogonal state in the $|\pm 1\rangle$ manifold. Therefore, we instead read out the spin state by applying an optical π pulse to the $|A_2\rangle$ state and collecting the emitted photons. We can choose to read out the population in a particular superposition of $|+1\rangle$ and $|-1\rangle$ by choosing the polarization of the readout laser. To separately characterize the effectiveness of this method, we first apply a magnetic field and use this technique to detect microwave-driven Rabi oscillations between $|+1\rangle$ and $|0\rangle$. Figure 2 shows a comparison of two methods for reading out the $|+1\rangle$ and $|0\rangle$ populations, along with the associated level scheme and pulse sequences. As expected, the oscillations are out of phase from each other. We observe that, compared to conventional resonant spin readout, the $|+1\rangle \rightarrow |A_2\rangle$ readout results in much fewer counts [26]. Nevertheless, we obtain good contrast and a signal-to-noise ratio of $\sim 11 : 1$ after about a minute of averaging.

We now use the combination of the spin polarization and readout techniques to demonstrate two-photon Rabi oscillations, as shown in Fig. 3. These experiments are conducted at

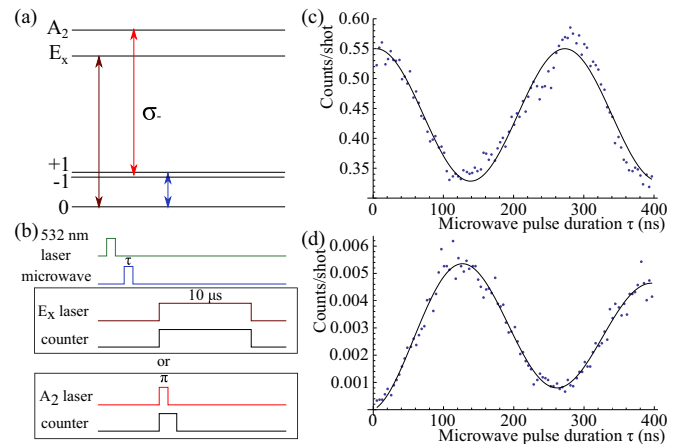


FIG. 2. (Color online) Readout of $|-1\rangle$ spin state population. (a) Level structure showing laser excitation and microwave field. A small external field of $B \sim 10 \text{ G}$ was applied. However, the Zeeman splitting is not resolvable with the $|+1\rangle \rightarrow |A_2\rangle$, so state selectivity is ensured by making the laser σ_- polarized. (b) Pulse sequence showing two alternative readout schemes. (c) Ground state Rabi oscillations detected using conventional spin readout with $|0\rangle \rightarrow |E_x\rangle$ transition. (d) Ground state Rabi oscillations detected using $|+1\rangle \rightarrow |A_2\rangle$ transition.

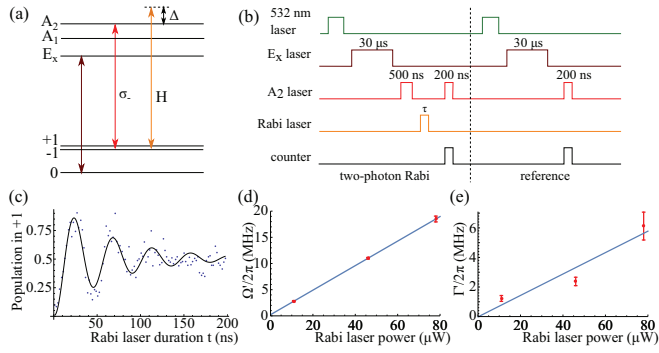


FIG. 3. (Color online) Two-photon Rabi oscillations between the $|\pm 1\rangle$ ground states. (a) Level structure showing the transitions involved. One laser is used for optical pumping from $|0\rangle$ state (magenta). Another σ_- polarized laser is used for optical pumping and spin readout from $|+1\rangle$ state (red). A third, linearly polarized laser is used for driving the two-photon transition (orange). (b) Pulse sequence showing spin preparation, manipulation, and readout steps. As a reference, the $|+1\rangle$ state was read out after applying only the $|0\rangle \rightarrow |E_x\rangle$ optical pumping step, which prepares an equal population in $|+1\rangle$ and $|-1\rangle$. (c) Two-photon Rabi oscillations with $46 \mu\text{W}$ of laser power. The counts obtained during the readout after two-photon Rabi are normalized to the counts obtained during readout after the reference sequence. (d) Two-photon Rabi frequency as a function of laser power, with linear fit (blue line). (e) Decay rate of two-photon Rabi oscillations as a function of laser power, with linear fit (blue line).

zero magnetic field, such that selective microwave excitation is not possible. First, the NV center is optically pumped into the $|-1\rangle$ state as described above. Using an EOM, we then apply a linearly polarized laser pulse of varying duration that is detuned $2\pi \times 2.24 \text{ GHz}$ from the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition. The population in the $|+1\rangle$ state is then measured with the same laser as the one used for $|+1\rangle$ to $|-1\rangle$ optical pumping. As can be seen in Fig. 3(c), we observe oscillations of the $|+1\rangle$ state population that eventually decays to a steady-state value of 1 relative to the reference, which corresponds to 50% population. Here, the two-photon Rabi frequency is large enough to drive all nuclear hyperfine levels of the ground state, which results in the much higher oscillation contrast compared to previous work [17]. As can be seen from Fig. 3(c), we achieve a π pulse in $\sim 25 \text{ ns}$ with a fidelity of $> 85\%$. While a fidelity of $> 99\%$ has been demonstrated using optimal control with microwaves, our π pulses are comparable in both speed and fidelity to conventional microwave manipulation [28].

In the case where the two branches of the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition are addressed by laser fields Ω_+ and Ω_- with large one-photon detuning Δ but zero two-photon detuning, the system behaves as if the $|\pm 1\rangle$ states are coupled by an effective two-photon Rabi frequency $\Omega' = \Omega_+^* \Omega_- / |\Delta|$. In our experiments, a single linearly polarized laser provides both circularly polarized driving fields. Two-photon Rabi oscillations occur in the limit where the detuning Δ is large enough so that $\frac{\Omega_+^2}{\Delta^2} \gamma, \frac{\Omega_-^2}{\Delta^2} \gamma \ll \Omega'$, where γ is the lifetime of the excited state. Additionally, one has to account for the presence of the $|\pm 1\rangle \rightarrow |A_1\rangle$ state with an additional detuning

of $2\pi \times 3.2 \text{ GHz}$. Since the $|A_1\rangle$ state has the opposite relative phase as the $|A_2\rangle$ state between the $|\pm 1\rangle$ spin components, we find that the two-photon Rabi frequency is given by

$$\Omega' = \Omega_+^* \Omega_- \left(\frac{1}{\Delta_2} - \frac{1}{\Delta_1} \right), \quad (1)$$

where $\Delta_{1,2}$ are the detunings from the $|A_{1,2}\rangle$ states, respectively. In Fig. 3(d), we perform a linear fit to the power dependence of Ω' . This allows us to extract the corresponding values of Ω_{\pm} , which are in good agreement with independently measured Rabi frequency of the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition.

The decay of the two-photon Rabi oscillations is due to several effects. First, for small two-photon Rabi frequencies, the hyperfine splitting due to the ^{14}N and ^{13}C spin bath results in decoherence of the Rabi oscillations. Second, off-resonant excitation leads to spontaneous emission from the excited state, leading to decay of the Rabi oscillations at a rate $\gamma(\Omega_+ \Omega_- / \Delta^2)$. Third, a combination of spectral diffusion of the $|A_2\rangle$ state and frequency fluctuations of the laser gives rise to an uncertainty in the detuning δ_Δ . This last effect is dominant in our case. The total decay rate of the two-photon Rabi oscillations is then given by

$$\Gamma = |\Omega_+| |\Omega_-| \left(\frac{1}{\Delta_2^2} - \frac{1}{\Delta_1^2} \right) \delta_\Delta. \quad (2)$$

Using this expression and the measured values of Ω' in Fig. 3(d), we extract a value of $\delta_\Delta = 2\pi \times 490 \text{ MHz}$ from a linear fit to the decay rates [Fig. 3(e)], consistent with independent optical spectroscopy measurements.

We next explore coherent transfer of population between the $|0\rangle$ and $|\pm 1\rangle$ states. This requires the coherent control of non-spin-preserving cross transitions between the ground and excited states [14,21]. We present spectroscopy and characterization of the tripod system formed between the $|A_2\rangle$ excited state and all three ground state levels in the same low-strain NV center as above. Remarkably, such a system allows for the possibility of nearly degenerate multiple dark resonances associated with transitions linking all different spin sublevels in the ground state manifold.

To demonstrate optically induced coherence between all three ground state spin levels, we scan a single linearly polarized laser across the $|0\rangle \rightarrow |A_2\rangle$ transition and modulate the same laser using an EOM to create sidebands for addressing the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition. We use a $50 \mu\text{s}$ excitation pulse and collect PSB photons during that interval. In addition, during each successive scan, we change the modulation frequency such that the sideband sweeps through the $|\pm 1\rangle \rightarrow |A_2\rangle$ transition. An external magnetic field was applied to split the $|\pm 1\rangle$ states by $2\pi \times 18 \text{ MHz}$. As can be seen in Fig. 4(c), when the sideband is far detuned from the $|\pm 1\rangle \rightarrow |A_2\rangle$ transitions, we simply obtain a resonance corresponding to the $|0\rangle \rightarrow |A_2\rangle$ cross transition. However, when the modulation frequency is such that the Λ systems involving the $|0\rangle$ and $|+1\rangle$ or $|0\rangle$ and $|-1\rangle$ states are in two-photon resonance, we observe a decrease in fluorescence corresponding to two nearly degenerate dark states.

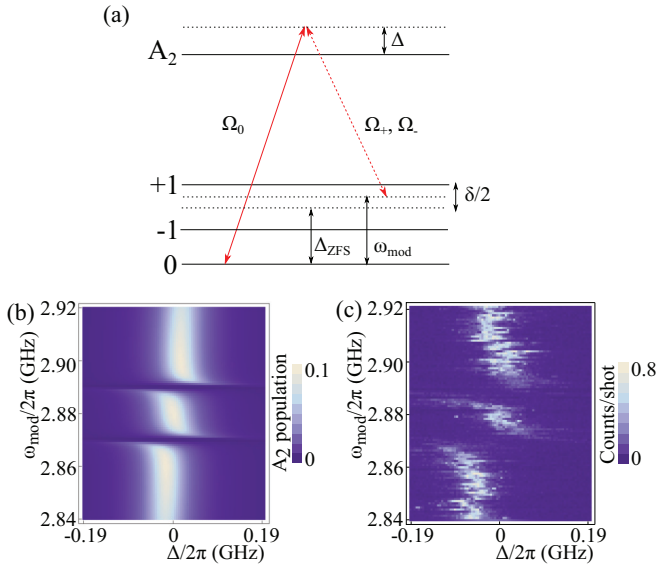


FIG. 4. (Color online) Observations of double-dark resonances in individual NV centers (a) Tripod level structure. (b) Theoretical excited state population as a function of one-photon detuning and modulation frequency. (c) Experimental data showing PSB fluorescence during laser excitation as a function of one-photon detuning and modulation frequency.

These observations can be understood by using a simple tripod model, described by the following Hamiltonian for the four-level system:

$$\begin{aligned}
 H = & -\Delta|0\rangle\langle 0| - \left(\Delta' + \frac{\delta}{2}\right)|+1\rangle\langle +1| \\
 & - \left(\Delta' - \frac{\delta}{2}\right)|-1\rangle\langle -1| \\
 & - (\Omega_0|A_2\rangle\langle 0| + \Omega_+|A_2\rangle\langle +1| + \Omega_-|A_2\rangle\langle -1| + \text{H.c.}). \quad (3)
 \end{aligned}$$

Here, as illustrated in Fig. 4(a), Δ is the one-photon detuning of the carrier laser frequency, δ is the Zeeman splitting between the $|\pm 1\rangle$ states, and $\Delta' = \Delta + \Delta_{\text{ZFS}} - \omega_{\text{mod}}$, where $\Delta_{\text{ZFS}} = 2\pi \times 2.88$ GHz is the ground state zero field splitting and ω_{mod} is the laser modulation frequency. Ω_0 is the Rabi frequency of the laser addressing the $|0\rangle \rightarrow |A_2\rangle$ transition. Importantly,

the two dark states associated with this model are

$$|D_{\pm}\rangle = \frac{\Omega_0|\pm 1\rangle - \Omega_{\pm}|0\rangle}{\sqrt{|\Omega_0|^2 + |\Omega_{\pm}|^2}}, \quad (4)$$

when $\Delta_{\text{ZFS}} - \omega_{\text{mod}} = \mp\delta/2$. The system stops absorbing light whenever either of these two dark resonance conditions is satisfied, regardless of the Zeeman splitting δ . This is the essence of double-dark resonances [18]. We can model the system including the effects of excited state decay using a full master equation approach. The results are shown in Fig. 4(b), and agree qualitatively with the experimental data. Remarkably, two nearly degenerate dark lines separated by the Zeeman splitting are observed, corresponding to double-dark resonances involving the states $|D_+\rangle$ and $|D_-\rangle$. Since the optical transitions between $|A_2\rangle$ and $|\pm 1\rangle$ have orthogonal circular polarizations, it is possible to achieve selective and independent coherent operations between $|0\rangle$ and either $|+1\rangle$ or $|-1\rangle$ even at zero magnetic field by choosing the appropriate laser detunings and polarizations.

The present observations open up new possibilities for the development of a full set of techniques for coherent control of the entire ground state manifold of the NV center using all-optical methods. We have shown that, using enhanced excitation and photon collection associated with a solid immersion lens, optical manipulation of the spin states can be achieved with comparable speed and fidelity to microwave manipulation. The four-level system investigated here can be used for coherent manipulation of all three ground states with, for example, the two-photon Rabi technique demonstrated here for the $|\pm 1\rangle$ states. Moreover, such a configuration enables a universal set of geometric gates on the $|\pm 1\rangle$ qubit states by using the $|0\rangle$ state as an ancilla [7,8]. The unique ability to selectively address a pair of dark resonances near degeneracy also present the possibility of studying novel interference effects and controlling the optical response [18]. These techniques can be immediately applicable to experiments involving NV centers in photonic structures such as optical cavities and waveguides [11,13,29]. With the development of diamond-based photonic devices that further enhance NV-light interactions, the techniques demonstrated can become important elements of integrated quantum network nodes based on NV centers in nanocavities.

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