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Generation of entangled photon strings using NV centers in diamond

D. D. Bhaktavatsala Rao, Sen Yang, and Jörg Wrachtrup
3. Physikalisches Institut, Research Center SCOPE, and MPI for Solid State Research, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

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We present a scheme to generate entangled photons using nitrogen vacancy (NV) centers in diamond. We show how the long-lived nuclear spin in diamond can mediate entanglement between multiple photons, thereby increasing the length of the entangled photon string. With the proposed scheme one could generate both n-photon Greenberger-Horne-Zeilinger and cluster states. We present an experimental scheme realizing the same and estimate the rate of entanglement generation both in the presence and absence of a cavity.

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With controlled generation and manipulation, quantum states of light are efficient carriers of quantum information with applications in quantum computing (QC), communications, and cryptography [1,2]. One of the major drawbacks with optical quantum information processing (QIP) is the absence of suitable nonlinear interactions to realize universal quantum gates, for example, a controlled-NOT (CNOT) gate between two photonic qubits. To overcome this difficulty, one may choose the implementation of desired quantum computation through a one-way quantum computer model [3] which requires the initialization of the quantum register in a globally entangled cluster state. The computation is then followed by performing only single qubit measurements. The one-way quantum computer or the measurement-based quantum computation using photonic qubits (polarization states) has already been shown to be a fault-tolerant model for QC and is tolerant to qubit losses [4]. The main hurdle in realizing optical QIP using this scheme is the generation of a multiqubit cluster state, the key initialization step of the model. While the experimental implementations succeeded to generate a six-qubit photonic cluster state optically [5], scaling this number further is not so clear. To this end there have been proposals to use solid-state emitters such as a periodically pumped quantum dot (QD) for the generation of cluster and Greenberger-Horne-Zeilinger (GHZ) states [6,7]. In this Rapid Communication we consider another possible solid-state system, the nitrogen vacancy (NV) centers in diamond, to generate multiphoton entangled states.

The NV center provides a hybrid spin system in which electron spins are used for fast [8], high-fidelity control [9] and readout [10,11], and nuclear spins are well isolated from their environment, yielding an ultralong coherence time [12]. Electron and nuclear spins could form a small-scale quantum register [13–15] allowing for, e.g., a necessary high-fidelity quantum error correction [13]. Furthermore, the NV electron spin can be entangled with an emitted optical photon [16,17] and further quantum entanglement [18] and quantum teleportation [19] between two remote NV centers have already been experimentally demonstrated. We have also recently demonstrated the ability of this solid-state device to store quantum information from a light field into the defect spins and a repetitive readout of the memory, essential for scalable networks. In addition, there have been other proposals to create large scalable QIP in diamond using a photonic architecture [20] where topological cluster state of the long-lived nuclear spins in various defect centers are generated using photons. Here we show the opposite, where the nuclear spin of a single defect center can mediate the entanglement between photons, thereby generating large strings of entangled photons.

The basic element of our system is a single NV center consisting of an electronic spin ($S=1$) and intrinsic ^{14}N nuclear spin ($I=1$), coupled by a hyperfine interaction. The interaction with optical photons in a Λ system forms the basis of our scheme, and is shown Fig. 1(a). The three-level Λ system is formed by the two ground states of the electron $\{|{+1}\rangle_e, |{-1}\rangle_e\}$ and an excited state $|A_{2(1)}\rangle$ [21]. Owing to the zero magnetic moment of the electron spin in the $|A_2\rangle$ state [21] and total angular momentum conservation, both ground states can be excited to the same state $|A_{2(1)}\rangle$ through absorption of a photon with σ_+ and σ_- polarization, respectively. We start with the two ground states being degenerate and the NV spin system prepared in the superposition state $|\Psi^+\rangle = \frac{1}{\sqrt{2}}(|{+1}\rangle_e + |{-1}\rangle_e)$. A photon in state $\frac{1}{\sqrt{2}}(|\sigma_+\rangle + |\sigma_-\rangle)$ and in resonance with the A_2 transition is sent into the NV center. After absorption of a photon, the collective photon-NV spin system is projected into the state $|A_2\rangle$, and hence the electron photon state after subsequent emission would remain in the entangled state, $|\psi_e^{(1)}\rangle = \frac{1}{\sqrt{2}}[|{+1}\rangle_e|\sigma_-\rangle + |{-1}\rangle_e|\sigma_+\rangle]$ [16,17]. Since the excitation process is as a projective measurement in the excited state basis, reexciting the NV system by a second photon pulse would immediately disentangle the first photon and the total state after the emission of the second photon would be $|\psi_e^{(2)}\rangle = \frac{1}{\sqrt{2}}[|{+1}\rangle_e|\sigma_-\rangle + |{-1}\rangle|\sigma_+\rangle] \otimes \frac{1}{\sqrt{2}}(|\sigma_+\rangle + |\sigma_-\rangle)$. Hence by postselecting only the absorption events one can see that the electron spin alone cannot mediate the entanglement between multiple photons, as found in other solid-state proposals [6]. To achieve this we use the hyperfine coupled nuclear spin to transfer the entanglement of the electron with the emitted photons to the nuclear spin, as detailed below.

After absorption and emission of the first photon the electron spin gets entangled to the photon $|\psi_e^{(1)}\rangle$, as described above. Before repumping the NV system with the second photon we perform a CNOT between the NV electron spin [see Fig. 1(b)] and its intrinsic ^{14}N spin, thus entangling them, viz., $|\psi_{en}^{(1)}\rangle = \frac{1}{\sqrt{2}}[|{+1}\rangle_e|\sigma_-\rangle|{+1}\rangle_n \pm |{-1}\rangle_e|\sigma_+\rangle|{-1}\rangle_n]$, where $|\pm 1\rangle_n$ are the basis states of the nuclear spin. A subsequent absorption and emission of the photon would leave the total system in the state, $|\psi_{en}^{(2)}\rangle = |\psi_e^{(2)}\rangle|\psi_n^{(1)}\rangle$, where the electron

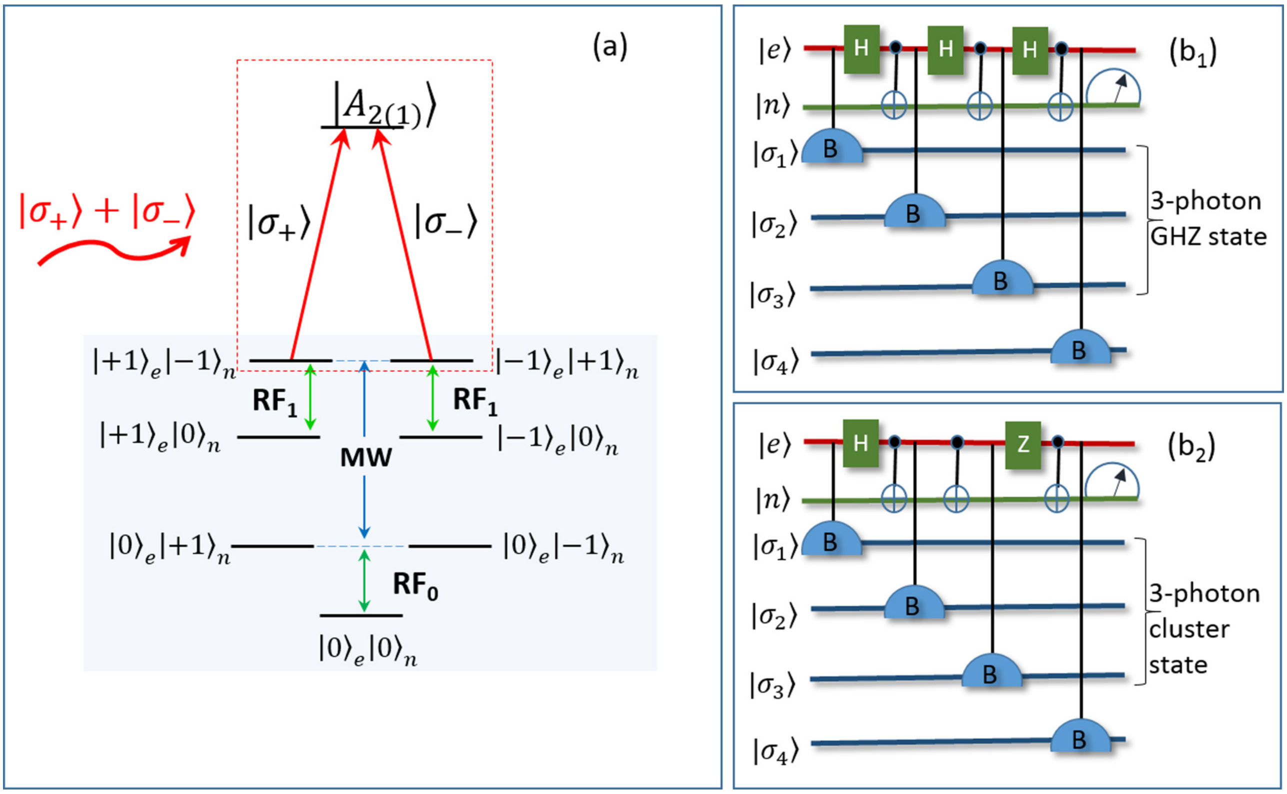

FIG. 1. (Color online) (a) The relevant level structure of the NV, with excited state $|A_{2(1)}\rangle = |E_-\rangle|+1\rangle_e \pm |E_+\rangle|-1\rangle_e$, and ground states $|E_0\rangle \otimes |m_s\rangle_e|m_n\rangle_n$, where $|E_{0,\pm}\rangle$ are orbital states with angular momentum projection $0, \pm1$, respectively. $|m_s = 0, \pm1\rangle_e$ and $|m_n = 0, \pm1\rangle_n$ are corresponding eigenstates of the electron spin and the ^{14}N nuclear spin. The spatial part of the wave function $|E_0\rangle$ is not explicitly written for simplicity when referring to ground states. The Λ system is highlighted. The individual manipulation of electron and nuclear spins by the microwave (MW) and radio-frequency (rf) fields is also shown in the figure. (b) Circuit diagram for generating a three-photon entangled state via the nuclear spin. H indicates the Hadamard gate on the electron spin in the two-level basis $|\pm\rangle_e$, and Z the standard controlled Pauli operations performed on the nuclear spin conditioned on the state of the electron. The interaction of the electron spin with the incoming photon is represented as a Bell type measurement (B) on the electron-photon system projecting it onto the entangled state $\frac{1}{\sqrt{2}}[|+1\rangle_e|\sigma_-\rangle + |-1\rangle_e|\sigma_+\rangle]$. Finally, a measurement on the nuclear spin will project the N-photon state onto the GHZ (b_1) or cluster state (b_2), as detailed in the text.

is now entangled with the second photon and the nuclear spin is entangled to the first photon. Following the circuit diagram in Fig. 1(b_1), and after the absorption of the third photon, the nuclear spin and the two photons are now projected on to the tripartite entangled state where the two photons are in the maximally entangled GHZ state for any spin projection of the nuclear spin, viz.,

$$|\phi_n^{(2)}\rangle = \frac{1}{\sqrt{2}}[|+1\rangle_n|G_-^{(2)}\rangle + |-1\rangle_n|G_+^{(2)}\rangle], \tag{1}$$

where $|G_\mp^{(2)}\rangle = \frac{1}{\sqrt{2}}[|\sigma_+\rangle|\sigma_+\rangle \mp |\sigma_-\rangle|\sigma_-\rangle]$. Continuing this procedure further, as shown in Fig. 1(b_1), an n-photon GHZ state is generated after the electron has been excited by a $(n+1)$th photon.

Instead, if we manipulate the electron spin with different local operations as shown in Fig. 1(b_2), we can, for example, project the nuclear spin and the two photons onto a different tripartite entangled state, as shown below after the absorption of the third photon:

$$|\phi_n^{(2)}\rangle = \frac{1}{\sqrt{2}}[|+1\rangle_n|C_0^{(2)}\rangle + |-1\rangle_n|C_1^{(2)}\rangle]. \tag{2}$$

Rewriting $|\sigma_\mp\rangle$ as $|0(1)\rangle$, $C_{0(1)}^{(2)}$, one can see that we have created a two-photon cluster state [22]

$$\begin{aligned} |C_0^{(2)}\rangle &= \frac{1}{2}\bigotimes_{a=1}^{2}(|0\rangle_a Z^{(a+1)} + |1\rangle_a), \\ |C_1^{(2)}\rangle &= \frac{1}{2}\bigotimes_{a=1}^{2}(|0\rangle_a + |1\rangle_a Z^{(a+1)}), \end{aligned} \tag{3}$$

where Z is the Pauli operator. Here we would like to highlight the possibility that by controlling the solid-state spins we could project the n-photon state onto different entangled states.

The proposed scheme can be implemented efficiently at low temperatures ($T < 8$ K) in a low strain ($\approx$1.2 GHz) NV center aligned along the [111] direction [16]. At such temperatures the optical transitions are well resolved, allowing for resonant excitation to perform efficient initialization and projective high-fidelity single shot readout on the electron spin. The degeneracy of the ground states can be maintained by switching off any external magnetic field. In addition, we can maintain the coherence of the nuclear spin for about a minute, a key parameter in our proposal. In solid-state proposals using the electron's spin, the rapid decoherence of the electronic spin could be a serious bottleneck for scalable production of entangled photons, which we could overcome in our present protocol. Due to the finite operation time for entangling each outgoing photon with the nuclear spin, there is a time delay τ between any two subsequent emissions. This immediately requires the emitted photon to be stored in a single mode optical fiber at the mode frequency of the zero phonon line (ZPL) of NV. Due to long T_1, the nuclear spin does not flip and hence the emitted photon remains entangled with it until the next photon arrives.

The next important factor to consider is the case when the electron spin is in the dark state to the applied laser field. To see this, for example, one can rewrite the entangled state of the total system, $\frac{1}{\sqrt{2}}[|+1\rangle_e|\phi^{(+1)}_{n,\sigma}\rangle + |-1\rangle_e|\tilde{\phi}^{(-1)}_{n,\sigma}\rangle]$, in the electron's bright and dark state basis ($|b(d)\rangle_e = \frac{1}{\sqrt{2}}[|+1\rangle_e \pm |-1\rangle_e]$), as $\frac{1}{\sqrt{2}}[|b\rangle_e|\psi^{(+1)}_{n,\sigma}\rangle + |d\rangle_e|\psi^{(-1)}_{n,\sigma}\rangle]$, where $\psi^{(\pm1)}_{n,\sigma}$ are the bright and dark combinations of $\phi^{(+1)}_{n,\sigma}$. Clearly there is a 50% probability of not having a resonant absorption at any time t. This makes the situation probabilistic as, for N photons incident on the NV center, the probability with which any m photons are resonantly absorbed has a Gaussian distribution $P(m) = \frac{N!}{(N-m)!(N+m)!} \approx \frac{2}{\sqrt{\pi N}} e^{-\frac{2(N-n)^2}{N}}$ centered around $N/2$ with width $\sqrt{N/2}$. Also, the m-photon state obtained for any two repetitive cycles of the protocol would be different, thereby reducing the fidelity of entanglement in the m-photon state.

To overcome this problem and only obtain an m-photon state consistently by only bright state absorptions, we switch on two lasers, allowing for an individual excitation on the transitions first to $|\pm1\rangle \leftrightarrow |A_1\rangle$, followed by the entangling transition $|\pm1\rangle \leftrightarrow |A_2\rangle$. If the electron is in the wrong state (dark state), then the $|A_1\rangle$ transition will populate it to the $|0\rangle_e$ state, thereby ending the operation cycle as no more absorption is possible (by this we eliminate the errors due to the dark state evolution in the presence of gate operations and hyperfine coupling that could mix it with the bright state). Instead, if the electron is in the correct state (bright state), it does not get excited to $|A_1\rangle$ and absorbs a photon resonant with the $|A_2\rangle$ transition. The probability of having m photons entangled at the end of an operation cycle will now depend on the probability distribution $P(m)$, which is given by

$$P_0(m) = \exp[-(m-1)\ln 2], \qquad (4)$$

for $N \gg m$. To estimate the event rate for generating entangled photons we shall define the time for an operation cycle $t = N\tau$ (τ is the separation between any two subsequent photons incident on the NV) and take a typical time scale $\tau = 1\ \mu$s. With an operation time of $t = 100\ \mu$s i.e., $N = 100$ photons are incident on the NV, there is a probability of 6.2×10^{-2} to have sequential absorption of five photons and a probability of 1.95×10^{-3} to have a sequential absorption of ten photons. Thus, under 1000 repetitions of the protocol, there will be ~62 events where the minimum length of the entangled chain of photons would be five, and two events with a minimum length of the entangled chain of photons being ten. These numbers are estimated for 100% collection efficiencies. For example, in the current experimental setup without a cavity, the probability to observe an entangled state with at least two photons is ~100 s. Such a low efficiency is also reported in the heralded entanglement with two NV's, where an entanglement event has been detected for every 2 min [18].

To get closer to the ideal estimate for the number of entangled photons, one can use a cavity to boost the emission into ZPL. For example, it was recently shown in an NV coupled photonic crystal cavity experiment [23] that, in the presence of a cavity, 70% of the emission would be in the ZPL, and with an achievable collection efficiency of ~90% [24] ZPL photons we achieve a ten-photon entangled state per second. A possible error that could decrease the entanglement fidelity when using a cavity is its imperfect circular symmetry that could reduce the degeneracy of the left and right circularly polarized light. This then leads to an additional dephasing of the photon entangled state. This dephasing should not be large when the polarization dependent cavity line shifts are within the linewidth of the ZPL. Alternatively, a polarization independent photonic cavity proposed in a recent work [25] could be a used to remove these additional dephasing effects.

The fidelity of generating an m-photon entangled state would decrease due to errors arising from imperfect gate operations and due to the dephasing of electron/nuclear spin coherence by the surrounding spin bath. During the protocol we perform multiple Hadamard and CNOT gate operations [see Fig. 1(b)] which entangle (mix) the basis states of the electron and nuclear spins. Any random phase obtained by these spins are eventually transferred into the m-photon state as the photons are always entangled to the nuclear spin [26]. Another source for this random phase is due to the spin noise caused by the quasistatic spin bath comprising the surrounding ^{13}C nuclear spins. In Fig. 2 we have plotted the fidelity $F_m = \mathrm{Tr}[\rho^{\mathrm{ideal}}_{m\sigma}\rho_{m\sigma}]$ as a function of m (entangled photons) in the presence of these common errors. Gate errors are introduced by imperfect unitary rotations that realize the Hadamard and CNOT gates, and the spin noise due to the coupling with (see

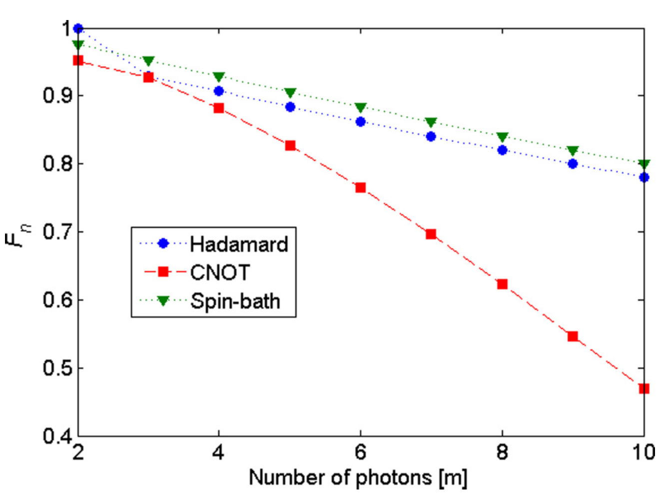

FIG. 2. (Color online) The fidelity of $F_m = \mathrm{Tr}[\rho^{\mathrm{ideal}}_{m\sigma}\rho_{m\sigma}]$ of the n-photon GHZ state generated by the solid-state spins is plotted as a function of m both in the presence of errors while performing the Hadamard and CNOT gates, and due to a random phase obtained by the nuclear spin at each interval τ due to the quasistatic spin bath. Both the rotation angle errors for the unitary gates and the phase error due to the spin bath are chosen to take a maximum value of 10°.

Ref. [26] for details) is included through a random phase obtained by the electron and nuclear spin during the operation t. Other dominant sources for nuclear spin dephasing could be due to the difference in the hyperfine coupling strength in the ground and excited states. This dephasing would be minimum in our case, as the excited levels $|A_{1(2)}\rangle$ are hardly perturbed by the hyperfine coupling [26] as opposed to other excited levels $E_{x,y}$ that are commonly used for initializing and readout of the electron spin [27].

In conclusion, we show that the long-lived nuclear spins in diamond could be a resource to mediate (generate) entanglement between single photons. The long $T_{1(2)}$ times of these spins allow for a controlled creation of multiphoton GHZ or cluster states, which could be useful for QIP with photons. Due to a weak coupling to the surrounding spin bath, random phase errors have less harmful effects than the errors in gate operations of the same order. We predicted the generation of an entangled photon string with a minimum length of ten photons per second in the presence of a cavity, where the emission of outgoing photons into the ZPL and the detection efficiencies are quite high. These solid-state spins, both with their ability to store quantum information from the incoming photons and generate entangled pairs, could have a promising impact on the field of quantum communications where on demand generation of high-fidelity entangled photons and their storage is quintessential.

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[1] P. Kok, W. J. Munro, K. Nemoto, T. C. Ralph, J. P. Dowling, and G. J. Milburn, Rev. Mod. Phys. **79**, 135 (2007).
[2] N. Gisin, G. Ribordy, W. Tittel, and H. Zbinden, Rev. Mod. Phys. **74**, 145 (2002).
[3] R. Raussendorf and H. J. Briegel, Phys. Rev. Lett. **86**, 5188 (2001).
[4] M. Varnava, D. E. Browne, and T. Rudolph, Phys. Rev. Lett. **97**, 120501 (2006).
[5] C.-Y. Lu *et al.*, Nat. Phys. **3**, 91 (2007).
[6] N. H. Lindner and T. Rudolph, Phys. Rev. Lett. **103**, 113602 (2009).
[7] S. E. Economou, N. Lindner, and T. Rudolph, Phys. Rev. Lett. **105**, 093601 (2010).
[8] G. D. Fuchs, V. V. Dobrovitski, D. M. Toyli, F. J. Heremans, and D. D. Awschalom, Science **326**, 1520 (2009).
[9] F. Dolde, V. Bergholm, Y. Wang, I. Jakobi, B. Naydenov, S. Pezzagna, J. Meijer, F. Jelezko, P. Neumann, T. Schulte-Herbruggen, J. Biamonte, and J. Wrachtrup, Nat. Commun. **5**, 3371 (2014).
[10] P. Neumann, J. Beck, M. Steiner, F. Rempp, H. Fedder, P. R. Hemmer, J. Wrachtrup, and F. Jelezko, Science **329**, 542 (2010).
[11] L. Robledo, L. Childress, H. Bernien, B. Hensen, P. F. Alkemade, and R. Hanson, Nature (London) **477**, 574 (2011).
[12] P. C. Maurer, G. Kucsko, C. Latta, L. Jiang, N. Y. Yao, S. D. Bennett, F. Pastawski, D. Hunger, N. Chisholm, M. Markham, D. J. Twitchen, J. I. Cirac, and M. D. Lukin, Science **336**, 1283 (2012).
[13] G. Waldherr, Y. Wang, S. Zaiser, M. Jamali, T. Schulte-Herbruggen, H. Abe, T. Ohshima, J. Isoya, J. F. Du, P. Neumann, and J. Wrachtrup, Nature (London) **506**, 204 (2014).
[14] F. Dolde, I. Jakobi, B. Naydenov, N. Zhao, S. Pezzagna, C. Trautmann, J. Meijer, P. Neumann, F. Jelezko, and J. Wrachtrup, Nat. Phys. **9**, 139 (2013).
[15] T. H. Taminiau, J. Cramer, T. van der Sar, V. V. Dobrovitski, and R. Hanson, Nat. Nanotechnol. **9**, 171 (2014).
[16] E. Togan, Y. Chu, A. S. Trifonov, L. Jiang, J. Maze, L. Childress, M. V. Dutt, A. S. Sorensen, P. R. Hemmer, A. S. Zibrov, and M. D. Lukin, Nature (London) **466**, 730 (2010).
[17] H. Kosaka and N. Niikura, Phys. Rev. Lett. **114**, 053603 (2015).
[18] H. Bernien, B. Hensen, W. Pfaff, G. Koolstra, M. S. Blok, L. Robledo, T. H. Taminiau, M. Markham, D. J. Twitchen, L. Childress, and R. Hanson, Nature (London) **497**, 86 (2013).
[19] W. Pfaff, B. J. Hensen, H. Bernien, S. B. van Dam, M. S. Blok, T. H. Taminiau, M. J. Tiggelman, R. N. Schouten, M. Markham, D. J. Twitchen, and R. Hanson, Science **345**, 532 (2014).
[20] K. Nemoto, M. Trupke, S. J. Devitt, A. M. Stephens, B. Scharfenberger, K. Buczak, T. Nöbauer, M. S. Everitt, J. Schmiedmayer, and W. J. Munro, Phys. Rev. X **4**, 031022 (2014).
[21] J. R. Maze, A. Gali, E. Togan, Y. Chu, A. Trifonov, E. Kaxiras, and M. D. Lukin, New J. Phys. **13**, 025025 (2011).
[22] H. J. Briegel and R. Raussendorf, Phys. Rev. Lett. **86**, 910 (2001).
[23] A. Faraon, C. Santori, Z. Huang, V. M. Acosta, and R. G. Beausoleil, Phys. Rev. Lett. **109**, 033604 (2012).
[24] L. Luozhou *et al.*, Nano Lett. **15**, 1493 (2015).
[25] B. K. H. Can,and M. G. Oner, IEEE Photonics Technol. Lett. **27**, 113 (2015).
[26] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.92.081301 for details on the effects of dephasing due to the nuclear spin bath, random phase errors, and state mixtures, and hyperfine interactions in the excited state.
[27] M. S. Blok, C. Bonato, M. L. Markham, D. J. Twitchen, V. V. Dobrovitski, and R. Hanson, Nat. Phys. **10**, 189 (2014).