Decoherence of Near-Surface Nitrogen-Vacancy Centers Due to Electric Field Noise

M. Kim,[1] H. J. Mamin,[1] M. H. Sherwood,[1] K. Ohno,[2,†] D. D. Awschalom,[2] and D. Rugar[1,*]
[1]*IBM Research Division, Almaden Research Center, San Jose, California 95120, USA*
[2]*Institute for Molecular Engineering, University of Chicago, Illinois 60637, USA*

(Received 21 May 2015; published 21 August 2015)

We show that electric field noise from surface charge fluctuations can be a significant source of spin decoherence for near-surface nitrogen-vacancy (NV) centers in diamond. This conclusion is based on the increase in spin coherence observed when the diamond surface is covered with high-dielectric-constant liquids, such as glycerol. Double-resonance experiments show that improved coherence occurs even though the coupling to nearby electron spins is unchanged when the liquid is applied. Multipulse spin-echo experiments reveal the effect of glycerol on the spectrum of NV frequency noise.

DOI: 10.1103/PhysRevLett.115.087602 PACS numbers: 76.30.Mi, 03.65.Yz, 05.40.Ca, 77.84.Nh

The negatively charged nitrogen-vacancy (NV) center in diamond is attracting great interest as an atomic-size quantum sensor that is operable at room temperature and has a convenient readout via optical fluorescence. NV centers are finding wide-ranging applications due to their responsiveness to local magnetic [1,2], electric [3,4], strain [5,6], and temperature fields [7,8]. In most cases, the sensitivity of the NV center is critically dependent on the long quantum coherence time of its spin state, which in bulk diamond can be greater than 1 ms at room temperature [9].

In many nanoscale sensing applications the NV center must be located as close to the surface as possible in order to maximize the detected signal [10–14]. Unfortunately, significant impairment of the spin coherence has been found for NV centers located within a few nanometers of the diamond surface [15–18]. In the NV-diamond research community, this near-surface decoherence is commonly attributed to magnetic noise emanating from unpaired electron spins in surface dangling bonds [15–19].

In this Letter we present evidence that near-surface NV decoherence is not solely due to magnetic noise, but instead can be dominated by electric field noise from surface charge fluctuations. This finding is based on the improvement of coherence seen when high-dielectric-constant liquids are applied to the diamond surface. For example, when the diamond is immersed in glycerol, we have found that Hahn-echo T_2 times can increase by more than a factor of 4. To rule out the influence of magnetic noise due to surface spins, we directly probed the surface electron spin density with a double-resonance experiment and found no significant change upon application of the glycerol. With simple electrostatic calculations, combined with the known NV spin Hamiltonian, we show that decoherence due to charge fluctuations is physically reasonable. Finally, we use the results from multipulse dynamic decoupling experiments to estimate the spectral density of the NV frequency noise.

Our experiments were performed using an electronic grade (100)-oriented diamond substrate that was capped with a 50-nm-thick layer of isotopically pure carbon-12 diamond. Near-surface NV centers were created by ^{15}N ion implantation at 2.5 keV, followed by annealing in vacuum at 850 °C, acid cleaning, and heating to 425 °C in a pure oxygen atmosphere [20]. This process results in NV centers located at depths roughly 5 nm below the surface. Individual NV centers were detected by confocal fluorescence microscopy with photon-counting electronics. The custom-built microscope had an inverted geometry that incorporated a small windowed cell which allowed liquid to be applied to the top surface of the diamond. See the Supplemental Material for further details on the sample preparation and apparatus [21].

Optically detected spin-echo experiments were performed with applied magnetic field in the range of 20–40 mT directed along the [111] symmetry axis of the NV center [Fig. 1(a)]. Measurements were made both before and after applying various liquids to the diamond surface. Four liquids were tested: conventional and fully deuterated glycerols (dielectric constant $\kappa_G = 42$), propylene carbonate ($\kappa_{PC} = 64$), and microscope immersion oil ($\kappa_{oil} = 2.3$). We note that glycerol and propylene carbonate have quite different chemical characteristics. Glycerol is an alcohol whose hydroxyl groups can donate protons to the environment, possibly leading to some passivation of surface dangling bonds. In contrast, propylene carbonate is known to be an aprotic solvent, meaning that the hydrogen atoms of the molecule are tightly bound.

As shown in Fig. 1(b), a dramatic 4.6-fold increase in T_2 was found when deuterated glycerol was placed on the diamond surface. Such a large increase indicates that the noise responsible for NV decoherence was substantially suppressed when the glycerol was added. After the glycerol was removed and the diamond recleaned, propylene carbonate was applied, again resulting in a significant 2.4-fold increase in T_2 time [Fig. 1(c)]. In contrast, when the same

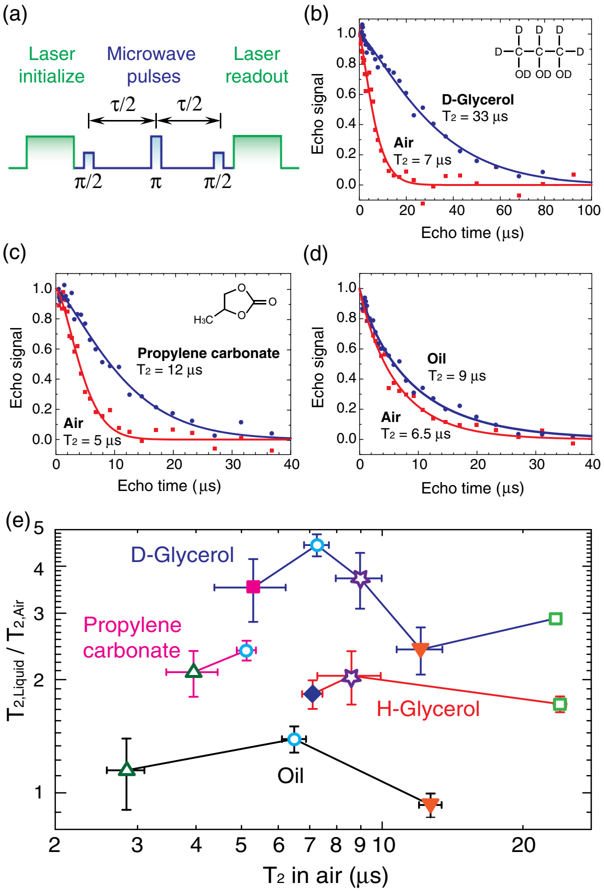

FIG. 1 (color online). Effect of various liquids on Hahn-echo T_2 times. (a) Pulse pattern for optically detected spin echo. (b)–(d) Normalized echo amplitudes obtained in air and with three different liquids covering the diamond surface. The same NV center was used for these three examples. Solid lines are fits to stretched exponentials. Bias field $B_z = 39$ mT. (e) Summary of T_2 ratios. The three liquids with high dielectric constant show a substantial increase in coherence time, with T_2 ratios between 1.7 and 4.6. Seven NV centers were tested, with each having a distinct symbol in the plot. Error bars are based on the standard error found from fitting the echo decays.

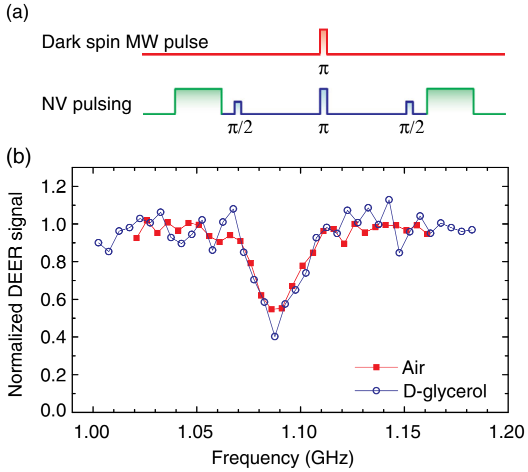

FIG. 2 (color online). DEER measurements. (a) Pulse pattern for the experiment. The frequency of the dark spin microwave (MW) pulse is scanned and causes spin inversions when the frequency matches the resonance frequency of the dark spins. The spin inversions are detected by their effect on the NV spin echo. (b) DEER measurements for a diamond sample in air and when covered with glycerol. The effect of the dark-spin inversions is clearly seen in the dip at 1.09 GHz. The addition of glycerol results in no substantial change in the dark-spin signal, indicating that glycerol does not significantly affect the dark-spin density. The echo evolution time was 5 μs.

NV center was studied with immersion oil, only a small 1.4-fold increase in T_2 was observed [Fig. 1(d)]. Similar comparisons were performed with six other NV centers, with results summarized in Fig. 1(e). Substantial improvements in NV coherence were found when any of the three high-κ liquids were applied to the diamond surface, with T_2 ratios ($T_{2,\mathrm{liquid}}/T_{2,\mathrm{air}}$) ranging from 1.7 to 4.6. In contrast, application of the lower-κ immersion oil showed little or no coherence improvement, with T_2 ratios ranging from 0.8 to 1.4.

To test whether the passivation of surface electron spins ("dark spins") is a possible mechanism of coherence improvement, we performed a double electron-electron resonance (DEER) experiment [Fig. 2(a)] [22,23]. We measured the NV spin echo while applying an additional microwave pulse halfway through the spin-echo sequence. This pulse inverts the dark spins when its frequency is resonant with the dark-spin precession frequency. The inversion of dark spins that are in close proximity to the NV causes a change the local magnetic field at the NV center and results in a dip in the echo response.

As can be seen in Fig. 2(b), a clear dip in the spin-echo signal occurs when the frequency of the microwave pulse matches the resonance frequency of the $g \approx 2$ dark spins (1.09 GHz in a 39 mT field), indicating that unpaired electron spins are indeed present in the neighborhood of the NV center. When the experiment was repeated after the addition of deuterated glycerol, the T_2 time increased by a factor of 2.4, but the DEER signal was essentially unchanged. Since no significant change is seen, it appears that the surface electron spin density is largely unaffected by the addition of the glycerol, and, thus, not the key factor in the observed T_2 improvement.

Given the DEER results above, we conclude that the improvement of coherence time with glycerol and propylene carbonate is most likely related to the high dielectric constants of these liquids, suggesting that much of the near-surface NV decoherence is the result of electric field noise due to fluctuating surface charges. A simple electrostatic calculation illustrates the action of the high-dielectric-constant liquid. Consider a point charge q on the surface of the diamond. The resulting electric field at the NV center depends on the dielectric constants of both the diamond and the external medium according to [24]

$$\mathbf{E} = \frac{1}{4\pi\varepsilon_0}\frac{2}{\kappa_d + \kappa_{\mathrm{ext}}}\frac{q}{r^2}\hat{\mathbf{r}}, \tag{1}$$

where r is the distance between the surface charge and the NV center, $\hat{\mathbf{r}}$ is the unit vector in the direction of the NV center, $\kappa_d = 5.7$ is the dielectric constant of diamond, κ_{ext} is the dielectric constant external to the diamond, and ε_0 is the permittivity of free space. Compared to a diamond in air, the reduction of electric field when an external medium (the liquid) is applied is given by $E/E_{\mathrm{air}} = (\kappa_d + 1)/(\kappa_d + \kappa_{\mathrm{ext}})$. For the case of glycerol with $\kappa_{\mathrm{ext}} = 42$, the electric field is thus reduced by a factor of 7. For a single electronic charge on the diamond surface, the electric field for a NV located 5 nm below the charge is 1.7×10^7 V/m when the diamond is in air, and reduced to 2.4×10^6 V/m with glycerol on the surface.

To show that fluctuating electric fields on the order of 10^7 V/m are sufficient to cause significant decoherence, we start with the NV spin Hamiltonian [3,25]

$$\begin{aligned} H = {} & (hD + d_\parallel E_z)[S_z^2 - 2/3] + \mu_B g_{\mathrm{NV}}\mathbf{S}\cdot\mathbf{B} \\ & - d_\perp[E_x(S_xS_y + S_yS_x) + E_y(S_x^2 - S_y^2)], \end{aligned} \tag{2}$$

where h is Planck's constant, $\mathbf{S}$ is the $S = 1$ electron spin operator, $\mathbf{B}$ is the applied magnetic field, $\mathbf{E}$ is the electric field at the NV center, $D = 2.87$ GHz is the zero field splitting, μ_B is the Bohr magneton, and $g_{\mathrm{NV}} \approx 2$ is the electron spin g factor. The electric field acts on the NV center via the coupling parameters $d_\parallel/h = 3.5\ \mathrm{mHz\,m\,V^{-1}}$ and $d_\perp/h = 170\ \mathrm{mHz\,m\,V^{-1}}$. To find the effect of electric field on the NV spin precession frequency, we assume the applied magnetic field is aligned with the NV symmetry axis (z axis). We can then solve for the energy eigenvalues associated with the three magnetic sublevels $m_s = +1$, 0, or -1 and find the precession frequencies for superpositions between the $m_s = 0$ and the ± 1 states. The resulting change in precession frequency due to an electric field is [3]

$$\Delta\omega_\pm/2\pi = (d_\parallel/h)E_z \pm \frac{1}{2}\frac{(d_\perp/h)^2E_\perp^2}{(\gamma/2\pi)B_z}, \tag{3}$$

where $E_\perp^2 = E_x^2 + E_y^2$, $\gamma/2\pi = g_{\mathrm{NV}}\mu_B/h = 28$ GHz/T, and we have assumed that $(d_\perp E_\perp/g_{\mathrm{NV}}\mu_B B_z)^2 \ll 1$.

With Eq. (3) we can now determine the frequency shift due to a single elementary charge located directly above a 5-nm-deep NV center. Assuming a magnetic field of 20 mT and a (100)-oriented diamond substrate, where the NV z-axis is tilted by 54.7° with respect to the surface normal, the E_z term contributes a 35-kHz shift. The $E_\perp$ term contributes an additional ±5 kHz, for a total frequency shift of up to 40 kHz. Frequency fluctuations of this magnitude would be sufficient to give a dephasing time $T_2^* \sim 1/|\Delta\omega|$ in the range of microseconds to tens of microseconds, depending on the spectrum of the fluctuations. While the effect of $E_\perp$ is fairly modest in this example, it becomes relatively more important the larger the electric field (i.e., when more charges are present and for shallower NVs), since it contributes quadratically in (3).

To better understand the frequency spectrum of the fluctuations that cause the near-surface decoherence, we performed multipulse dynamic decoupling experiments [Fig. 3(a)] [16,18,26,27]. NVs were studied both before and after the application of deuterated glycerol using $XY8$-N pulse sequences [11,28], where N is the number of π pulses in the sequence ($N = 1$, 32, 96, and 256). Figures 3(b) and 3(c) show spin coherence data as a function of total evolution time based on measured spin-echo amplitudes. The curves were found to be well fit by

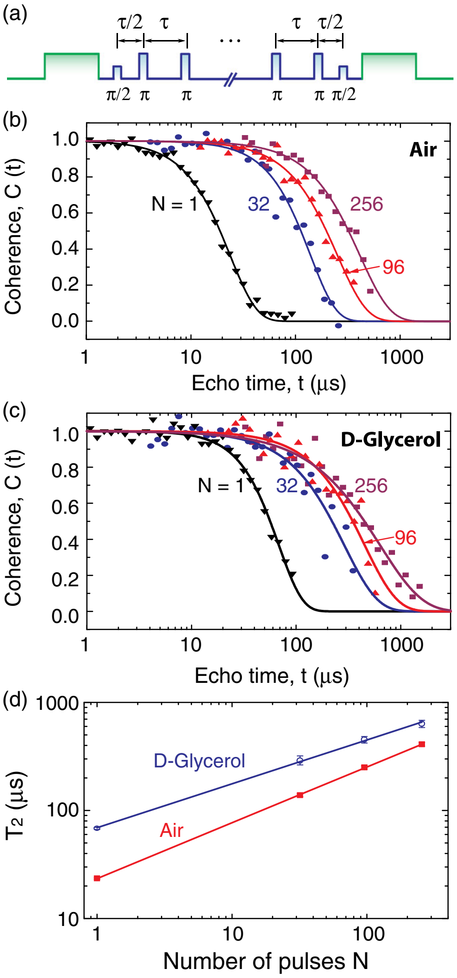

FIG. 3 (color online). Results of multipulse spin-echo measurements. (a) Pulse sequence for the measurements. The π pulse phases were in an $XY8$-N pattern, where N is the number of π pulses. (b) Points are coherence data (normalized spin-echo amplitudes) taken in air as a function of echo time $t = N\tau$ with a bias field $B_z = 21$ mT. Solid curves are fits to stretched exponentials. (c) Same as for panel (b) but measured with deuterated glycerol covering the diamond. (d) T_2 as a function of number of π pulses. T_2 is proportional to $N^{0.52}$ in air and $N^{0.41}$ in deuterated glycerol.

stretched exponentials of the form $\exp[-(t/T_2)^n]$. As expected, the T_2 times increased with the number of π pulses [Fig. 3(d)], and exhibited the power-law dependence $T_2 \propto N^k$, with $k = 0.52$ in the case of air and 0.41 in that with glycerol.

The coherence data in Fig. 3 can be used to estimate the spectrum of NV frequency fluctuations by taking into account the filter functions associated with the decoupling sequences [29]. Using a spectral decomposition procedure similar to that described in Refs. [18] and [27], we extract an estimate for the spectral density of the NV precession frequency, $S_\omega(\omega)$ [30]. In air, the spectrum roughly fits a $1/\omega$ dependence between 10 kHz and 1 MHz (Fig. 4). The addition of glycerol substantially reduces the spectral density for frequencies between 10 kHz and 100 kHz, where a dependence of $\sim 1/\omega^{0.8}$ is seen. It is this reduction of spectral density that is most responsible for the observed increase in T_2 times. Above 100 kHz, the glycerol spectrum flattens out, and beyond 600 kHz, the spectral density with glycerol is approximately equal to the spectral density without glycerol.

The ineffectiveness of glycerol to cancel electric field noise above 600 kHz is somewhat surprising given that the dielectric relaxation frequency for bulk liquid glycerol has been measured to be greater than 100 MHz [31]. One possibility is that the dielectric relaxation frequency is much reduced at the surface of the diamond. For example, experiments probing nanoscale layers of glycerol on surfaces have found evidence that a nanometer-thick layer of reduced mobility can form at the solid-liquid interface [32–34]. This semisolid layer could impede the rotation of glycerol molecules and thereby reduce the effective dielectric constant at high frequencies. A second possibility is that thermal agitation of the glycerol molecules adds broadband electric field noise and thereby sets a floor to the spectral density that becomes the dominant noise source at higher frequencies. A straightforward calculation shows that randomly rotating electric dipoles from glycerol molecules will create a substantial fluctuating electric field of approximately 10^7 V/m rms at a depth of 5 nm.

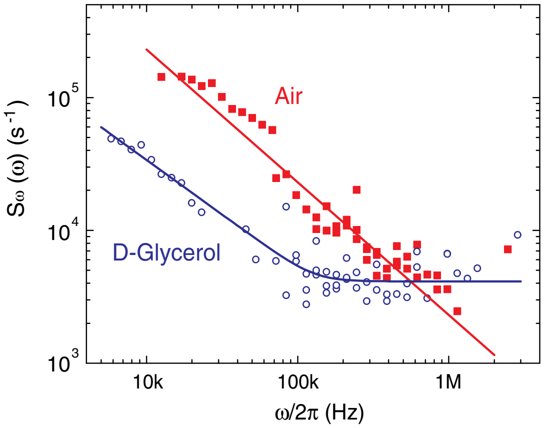

FIG. 4 (color online). Power spectral density of precession frequency noise as determined from spectral decomposition of multipulse coherence data. In air, the spectral density falls roughly as $1/\omega$, indicated by the solid red line. In deuterated glycerol, the response is approximately $1/\omega^{0.8}$ for frequencies below 100 kHz and levels off for higher frequencies. The solid blue line is a guide for the eye.

It is tempting to use our estimate of $S_\omega(\omega)$ to find the electric field spectral density. Unfortunately, with our current data set, the frequency-mixing behavior of the $E_\perp^2$ nonlinearity in (3) makes it impossible to rigorously determine the electric field spectral densities without making some significant assumptions about the noise spectrum in frequency regions where we have no direct experimental information. For example, a substantial dc electric field from static surface charge would not be directly evident in our measurements, but would act to enhance the relative contribution of fluctuating fields via the $E_\perp^2$ nonlinearity.

If we take a naïve approach and consider only the E_z contribution in (3), then the analysis is straightforward and we can write $S_\omega = 4\pi^2(d_\parallel/h)^2 S_{E_z}$. To find the electric field spectral density S_{E_z} we take S_ω in air from Fig. 4, which is approximately $S_\omega = 1.4 \times 10^{10}\mathrm{s}^{-2}/|\omega|$, and obtain $S_{E_z} = 2.9 \times 10^{13}(\mathrm{V/m})^2/|\omega|$. Integrating this over the measured range of 10 kHz to 1 MHz, we find $\langle E_z^2\rangle^{1/2} = 6.5 \times 10^6$ V/m, which is less than the equivalent of one electronic charge at 5-nm distance. This value should be viewed as a very conservative lower bound to the total fluctuating field, since we are considering only one vector component of field and over a very limited frequency range.

In closing, we note that an alternative approach for distinguishing between electric and magnetic field noise in NV decoherence is to compare conventional spin-echo results with "double-quantum" spin echoes, which utilize the superposition between the $m_s = -1$ and $+1$ sublevels [35–38]. We explore this avenue in the Supplemental Material [21] and show results that support our conclusion that electric field noise can be a significant contributor to decoherence for near-surface NV centers.

The authors thank B. Myers, A. Jayich, M. Salmeron, and J. Hodges for helpful discussions. This work was supported by the DARPA QuASAR program and the U.S. Air Force Office of Scientific Research.

---

[*] Correspondence author.
rugar@us.ibm.com

[†] Present address: Applied Materials, Inc., 3340 Scott Boulevard, Santa Clara, California 95054, USA.

[1] G. Balasubramanian, I. Y. Chan, R. Kolesov, M. Al-Hmoud, J. Tisler, C. Shin, C. Kim, A. Wojcik, P. R. Hemmer, A. Krueger, T. Hanke, A. Leitenstorfer, R. Bratschitsch, F. Jelezko, and J. Wrachtrup, Nature (London) **455**, 648 (2008).

[2] J. R. Maze, P. L. Stanwix, J. S. Hodges, S. Hong, J. M. Taylor, P. Cappellaro, L. Jiang, M. V. G. Dutt, E. Togan, A. S. Zibrov, A. Yacoby, R. L. Walsworth, and M. D. Lukin, Nature (London) **455**, 644 (2008).

[3] F. Dolde, H. Fedder, M. W. Doherty, T. Nöbauer, F. Rempp, G. Balasubramanian, T. Wolf, F. Reinhard, L. C. L. Hollenberg, F. Jelezko, and J. Wrachtrup, Nat. Phys. **7**, 459 (2011).

[4] F. Dolde, M. W. Doherty, J. Michl, I. Jakobi, B. Naydenov, S. Pezzagna, J. Meijer, P. Neumann, F. Jelezko, N. B. Manson, and J. Wrachtrup, Phys. Rev. Lett. **112**, 097603 (2014).

[5] P. Ovartchaiyapong, K. W. Lee, B. A. Myers, and A. C. B. Jayich, Nat. Commun. **5**, 4429 (2014).

[6] E. R. MacQuarrie, T. A. Gosavi, N. R. Jungwirth, S. A. Bhave, and G. D. Fuchs, Phys. Rev. Lett. **111**, 227602 (2013).

[7] D. M. Toyli, C. F. de las Casas, D. J. Christle, V. V. Dobrovitski, and D. D. Awschalom, Proc. Natl. Acad. Sci. U.S.A. **110**, 8417 (2013).

[8] G. Kucsko, P. C. Maurer, N. Y. Yao, M. Kubo, H. J. Noh, P. K. Lo, H. Park, and M. D. Lukin, Nature (London) **500**, 54 (2013).

[9] G. Balasubramanian, P. Neumann, D. Twitchen, M. Markham, R. Kolesov, N. Mizuochi, J. Isoya, J. Achard, J. Beck, J. Tissler, V. Jacques, P. R. Hemmer, F. Jelezko, and J. Wrachtrup, Nat. Mater. **8**, 383 (2009).

[10] H. J. Mamin, M. Kim, M. H. Sherwood, C. T. Rettner, K. Ohno, D. D. Awschalom, and D. Rugar, Science **339**, 557 (2013).

[11] T. Staudacher, F. Shi, S. Pezzagna, J. Meijer, J. Du, C. A. Meriles, F. Reinhard, and J. Wrachtrup, Science **339**, 561 (2013).

[12] M. S. Grinolds, S. Hong, P. Maletinsky, L. Luan, M. D. Lukin, R. L. Walsworth, and A. Yacoby, Nat. Phys. **9**, 215 (2013).

[13] D. Rugar, H. J. Mamin, M. H. Sherwood, M. Kim, C. T. Rettner, K. Ohno, and D. D. Awschalom, Nat. Nanotechnol. **10**, 120 (2015).

[14] T. Häberle, D. Schmid-Lorch, F. Reinhard, and J. Wrachtrup, Nat. Nanotechnol. **10**, 125 (2015).

[15] T. Rosskopf, A. Dussaux, K. Ohashi, M. Loretz, R. Schirhagl, H. Watanabe, S. Shikata, K. M. Itoh, and C. L. Degen, Phys. Rev. Lett. **112**, 147602 (2014).

[16] B. A. Myers, A. Das, M. C. Dartiailh, K. Ohno, D. D. Awschalom, and A. C. Bleszynski Jayich, Phys. Rev. Lett. **113**, 027602 (2014).

[17] B. K. Ofori-Okai, S. Pezzagna, K. Chang, M. Loretz, R. Schirhagl, Y. Tao, B. A. Moores, K. Groot-Berning, J. Meijer, and C. L. Degen, Phys. Rev. B **86**, 081406(R) (2012).

[18] Y. Romach, C. Müller, T. Unden, L. J. Rogers, T. Isoda, K. M. Itoh, M. Markham, A. Stacey, J. Meijer, S. Pezzagna, B. Naydenov, L. P. McGuinness, N. Bar-Gill, and F. Jelezko, Phys. Rev. Lett. **114**, 017601 (2015).

[19] L. Luan, M. S. Grinolds, S. Hong, P. Maletinsky, R. L. Walsworth, and A. Yacoby, Sci. Rep. **5**, 8119 (2015).

[20] M. Kim, H. J. Mamin, M. H. Sherwood, C. T. Rettner, J. Frommer, and D. Rugar, Appl. Phys. Lett. **105**, 042406 (2014).

[21] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.115.087602 for details on methods, apparatus, and supporting experiments.

[22] B. Grotz, J. Beck, P. Neumann, B. Naydenov, R. Reuter, F. Reinhard, F. Jelezko, J. Wrachtrup, D. Schweinfurth, B. Sarkar, and P. Hemmer, New J. Phys. **13**, 055004 (2011).

[23] H. J. Mamin, M. H. Sherwood, and D. Rugar, Phys. Rev. B **86**, 195422 (2012).

[24] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975), pp. 147–149.

[25] E. Van Oort and M. Glasbeek, Chem. Phys. Lett. **168**, 529 (1990).

[26] G. de Lange, Z. H. Wang, D. Ristè, V. V Dobrovitski, and R. Hanson, Science **330**, 60 (2010).

[27] N. Bar-Gill, L. M. Pham, C. Belthangady, D. Le Sage, P. Cappellaro, J. R. Maze, M. D. Lukin, A. Yacoby, and R. Walsworth, Nat. Commun. **3**, 858 (2012).

[28] T. Gullion, D. B. Baker, and M. S. Conradi, J. Magn. Reson. **89**, 479 (1990).

[29] Ł. Cywinski, R. M. Lutchyn, C. P. Nave, and S. Das Sarma, Phys. Rev. B **77**, 174509 (2008).

[30] We assume a double-sided convention for the spectral density with the normalization $(1/2\pi)\int_{-\infty}^{\infty} S_\omega(\omega)d\omega = \langle(\Delta\omega)^2\rangle$, where $\langle(\Delta\omega)^2\rangle$ is the mean square of the frequency fluctuations.

[31] K. L. Ngai, P. Lunkenheimer, C. León, U. Schneider, R. Brand, and A. Loidl, J. Chem. Phys. **115**, 1405 (2001).

[32] S. Capponi, S. Napolitano, N. R. Behrnd, G. Couderc, J. Hulliger, and M. Wübbenhorst, J. Phys. Chem. C **114**, 16696 (2010).

[33] L. Xu and M. Salmeron, J. Phys. Chem. B **102**, 7210 (1998).

[34] With both glycerol and propylene carbonate we have frequently seen the T_2 improvement diminish over the course of several hours. We attribute this to interfacial solidification, which will reduce the effective dielectric constant.

[35] N. Zhao, Z.-Y. Wang, and R.-B. Liu, Phys. Rev. Lett. **106**, 217205 (2011).

[36] P. Huang, X. Kong, N. Zhao, F. Shi, P. Wang, X. Rong, R.-B. Liu, and J. Du, Nat. Commun. **2**, 570 (2011).

[37] K. Fang, V. M. Acosta, C. Santori, Z. Huang, K. M. Itoh, H. Watanabe, S. Shikata, and R. G. Beausoleil, Phys. Rev. Lett. **110**, 130802 (2013).

[38] H. J. Mamin, M. H. Sherwood, M. Kim, C. T. Rettner, K. Ohno, D. D. Awschalom, and D. Rugar, Phys. Rev. Lett. **113**, 030803 (2014).