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ABSTRACT
Quantum sensors based on the electron spin states of nitrogen vacancy centers in diamond have found wide application in magnetometry and nuclear spin magnetic resonance measurements. Previously, we have theoretically and experimentally investigated the effect of finite pulse width on quantum sensing for synchronous alternating-current (AC) magnetic fields to dynamical decoupling sequences [T. Ishikawa et al., Phys. Rev. Appl. 10, 054059 (2018)]. However, many biological and condensed-matter systems exhibit fluctuating AC fields over time, and thus, our model needs modifications because of additional non-ideal conditions in practical measurements. Here, we investigate the effects of finite pulse width of multiple-pulse decoupling sequences on quantum sensing for asynchronous AC magnetic fields. For this purpose, we use a spin ensemble of nitrogen vacancy centers in an isotopically purified diamond film. We reveal through experiments that the finite-width pulse causes shifts in AC magnetometry signals in a free-precession-time plot. In addition, our results indicate that the finite-width pulse affects the amplitude of magnetometry signals, implying that the finite-pulse-width effect should be taken into account for realizing accurate measurement of the frequency and amplitude of asynchronous AC magnetic fields.

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At present, highly sensitive magnetic field sensors are widely used not only for fundamental research on magnetism and spin dynamics but also for practical applications such as medical imaging based on nuclear spin magnetic resonance (NMR). Among such sensors, a quantum sensor based on the electron spin states of a nitrogen vacancy (NV) center in diamond, i.e., a qubit, has gained considerable attention in magnetometry and NMR applications owing to its high sensitivity and a nanometer-scale resolution. For these applications, the amplitude of an alternating-current (AC) magnetic field is generally measured by a qubit operating with dynamic decoupling sequences having multiple pulses (hereafter referred to as multiple-pulse decoupling sequences). The maximum phase accumulation for the qubit can be attained from an AC magnetic field when its frequency matches that of multiple-pulse decoupling sequences and the relative phase between the AC field and the sequences is zero. This phase-accumulation condition can enhance the signals from a particular frequency field and suppress unwanted frequency signals or noise. Therefore, a qubit with multiple-pulse decoupling sequences can serve as a lock-in amplifier in the quantum regime, which can realize sensitive measurements of AC magnetic fields with small amplitudes.

Multiple-pulse decoupling sequences can also improve the coherence time of a qubit, and thus its frequency resolution and sensitivity. Characterizations of multiple-pulse decoupling...
sequences are generally modeled by filter functions with infinitely narrow pulses for time evolution of spin states. However, as we have previously discussed, because pulses possess finite width in practice, the filter functions should be modified. In our previous work, we proposed a model of finite-pulse-width effect on AC magnetometry by using multiple-pulse decoupling sequences under a synchronous condition in which the phase difference between an AC magnetic field and a decoupling sequence is zero. Using the proposed model, we demonstrated the detection of an AC magnetic field with an accurate frequency and linewidth in quantum sensing with multiple-pulse decoupling sequences. As well as our previous work, AC magnetometry for the synchronous fields is well exploited for demonstrating the proof-of-concept of quantum sensing. In addition, quantum sensing for the synchronous fields to the decoupling sequences is also researched for the measurements of current-induced magnetic fields in micrometer and nanometer scales and NMR measurements of polarized nuclear spins. However, many biological and condensed-matter systems exhibit fluctuating magnetic fields over time, and thus, our model about the pulse width effect still needs modifications because of additional non-ideal conditions in practical measurements. For example, in practical AC magnetometry of unpolarized nuclear spins, a spin in the NV center (qubit) is located under AC magnetic fields with random phases. In such a case, the phase differences between the AC magnetic fields and the decoupling sequences are randomly distributed in the range 0–2π; therefore, AC magnetometry signals by using decoupling sequences need to be modified. In fact, the AC magnetometry signals based on the decoupling sequences are proportional to the zeroth-order Bessel function of the first kind $J_0(\Phi(B_{ac}, f_{ac}))$ for AC magnetic fields with randomly distributed phases (i.e., asynchronous AC magnetic fields), in contrast to sin $\Phi$ (or cos $\Phi$) for synchronous AC magnetic fields, where $B_{ac}$ and $f_{ac}$ are the amplitude and frequency of AC magnetic fields, respectively. However, the effects of finite pulse width have not yet been investigated for AC magnetometry signals of asynchronous AC magnetic fields. In this study, therefore, we investigated the effects of finite pulse width on AC magnetometry for asynchronous AC magnetic fields and verified the model by AC magnetometry experiments using NV centers in diamond.

We used a spin ensemble of NV centers ([NV] = 3 × 10$^{14}$ cm$^{-3}$) in an isotopically purified diamond film of thickness ≈50 nm. An AC magnetic field was continuously applied to the NV-spin ensemble by a home-made coil connected to an arbitrary function generator (Tektronix AFG3252), keeping the AC magnetic field asynchronous to the decoupling sequence. The sample and the experimental setup are the same as those used in our previous works.

In this study, we applied an AC magnetic field given by $B(t) = B_{ac}\cos(\omega_{ac}t + \phi_{ac})$, where $B_{ac}$ is the field amplitude and $\phi_{ac}$ is the initial phase at the start of the decoupling sequences, which corresponds to the phase difference between the AC magnetic field and the decoupling sequence. $\omega_{ac} = 2\pi f_{ac}$, where $f_{ac}$ is the frequency of the AC field. Phase accumulation due to an AC magnetic field synchronous to a Hahn-echo sequence is given by

$$\Phi_{\text{echo}} = \frac{4y_{NV}B_{ac}}{\omega_{ac}} \sin\left(\frac{\omega_{ac}\tau(1 + \alpha)}{4}\right) \sin\left(\frac{\omega_{ac}\tau(1 + 2\alpha)}{4}\right),$$

where $\tau$ is the free precession time [Fig. 1(a)]; $y_{NV}$ is the gyromagnetic ratio of the electron spin of an NV center, given by $y_{NV}/2\pi = 28$ GHz/T; and $\alpha$ is the ratio of $\pi$ pulse width $\tau_p$ to $\tau$, i.e., $\tau_p = \alpha \tau$. The AC magnetometry signal is given by $\cos \Phi_{\text{echo}}$ when the first $\pi/2$ pulse is in phase with that of the last $\pi/2$ pulse, as presented in Fig. 1(a). To examine AC magnetometry for an asynchronous AC magnetic field by using the Hahn-echo sequence, we assumed that $\phi_{ac}$ in Eq. (1) is uniformly random-distributed in the range of 0–2π and acquired the averaged magnetometry signal over $\phi_{ac} = 0–2\pi$, which is given by

$$S_{\text{echo}} = \langle \cos \Phi_{\text{echo}} \rangle_{\phi_{ac}} e^{-\tau/T_S},$$

$$= J_0\left(\frac{4y_{NV}B_{ac}}{\omega_{ac}} \sin\left(\frac{\omega_{ac}\tau(1 + \alpha)}{4}\right) \sin\left(\frac{\omega_{ac}\tau(1 + 2\alpha)}{4}\right)\right) e^{-\tau/T_S},$$

Here, $\langle \rangle_{\phi_{ac}}$ denotes averaging over $\phi_{ac}$ and $J_0$ is the zeroth order Bessel function of the first kind. The Bessel term in Eq. (2) is unity as $B_{ac} = 0$. Therefore, the baseline of Hahn-echo magnetometry signals is subjected to exponential decay due to decoherence of spin states in the NV centers, characterized by coherence time $T_2$. We measured $T_2$ of the NV-spin ensemble by using the Hahn-echo sequence without an AC magnetic field and obtained $T_2 = 74 \pm 3$ μs and $p = 0.95 \pm 0.04$. $S_{\text{echo}} \propto J_0\left[2y_{NV}B_{ac} \sin^2(\pi f_{ac}\tau/2)/\pi f_{ac}\right]$ if $\alpha = 0$, which is consistent with Ref. 26.

Figure 1(b) shows the AC magnetometry results as a function of free precession time $\tau$ for the Hahn-echo sequences with the following conditions: 14 cm$^{-1}$ AC magnetic field, $\pi$ pulse width $\tau_p = 34\mu s$, and $\pi/2$ pulse width $\tau_{\pi/2} = 14\mu s$. As shown in the figure, the baseline of magnetometry signal was observed due to the exponential decay. The dependence of magnetometry signals on $\tau$ is proportional to the zeroth-order Bessel function of the first kind $J_0(\Phi(B_{ac}, f_{ac}))$ and $\cos \Phi_{\text{echo}}$, as shown in Eq. (2). The AC magnetometry signal is given by $\cos \Phi_{\text{echo}}$ when the first $\pi/2$ pulse is in phase with that of the last $\pi/2$ pulse, as presented in Fig. 1(a). To examine AC magnetometry for an asynchronous AC magnetic field by using the Hahn-echo sequence, we assumed that $\phi_{ac}$ in Eq. (1) is uniformly random-distributed in the range of 0–2π and acquired the averaged magnetometry signal over $\phi_{ac} = 0–2\pi$, which is given by

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Here, $\langle \rangle_{\phi_{ac}}$ denotes averaging over $\phi_{ac}$ and $J_0$ is the zeroth order Bessel function of the first kind. The Bessel term in Eq. (2) is unity as $B_{ac} = 0$. Therefore, the baseline of Hahn-echo magnetometry signals is subjected to exponential decay due to decoherence of spin states in the NV centers, characterized by coherence time $T_2$. We measured $T_2$ of the NV-spin ensemble by using the Hahn-echo sequence without an AC magnetic field and obtained $T_2 = 74 \pm 3$ μs and $p = 0.95 \pm 0.04$. $S_{\text{echo}} \propto J_0\left[2y_{NV}B_{ac} \sin^2(\pi f_{ac}\tau/2)/\pi f_{ac}\right]$ if $\alpha = 0$, which is consistent with Ref. 26.
\(\pi\)-pulse width \(\tau_s = 124\) ns (red circles), 374 ns (blue circles), and 622 ns (green circles). For all measurements, we set \(f_{ac} = 500\) kHz and \(B_{ac} = 4.5\) \(\mu T\). Note that the \(B_{ac}\) value was evaluated from the data fitting results (see later). To simulate the measurement under AC fields with randomly distributed \(\phi_{ac}\) values, AC magnetometry measurements were repeatedly performed by randomly varying \(\phi_{ac}\) for each \(\pi\) pulse width. In Fig. 1(b), repetition times were \(2 \times 10^6\), \(1 \times 10^5\) and \(5 \times 10^4\) for the AC magnetometry measurements with the \(\pi\) pulse width \(\tau_s = 124\), 374 ns and 624 ns, respectively. Each experimental result shown in Fig. 1(b) is the average value of the data obtained from the repeated measurements using each \(\tau_s\) condition. Note that the \(\pi\) pulse width \(\tau_s = 124\) ns, 374 ns, and 622 ns correspond to \(\pi\), \(3\pi\), and \(5\pi\), respectively. Figure 1(b) also shows the calculated result of AC magnetometry (black dashed line) for \(\tau_s = 0\) (\(\alpha = 0\)), i.e., the formula given in Ref. 26. In this case, resonance signals appear around \(\tau = k f_{ac}\) (\(k = 1, 3, 5, \cdots\)). By contrast, resonance signals are observed at lower \(\tau\) values in the experiments for \(\tau_s \neq 0\). This result can be understood from Eq. (2) with finite \(\alpha\) values. According to Eq. (2), resonance signals appear at \(\tau = k f_{ac}(1 + \alpha)\) due to the second sinusoidal term in the Bessel function. In fact, for \(\tau_s = 124\) ns, the two resonance signals are observed around \(\tau = 1.876\) \(\mu s\) and 5.876 \(\mu s\), which coincide with \(k f_{ac}(1 + \alpha)\) with \(k = 1\) and \(k = 3\), respectively. Note that the positions of resonance signals for other \(\tau_s\) conditions also coincide with \(\tau = k f_{ac}(1 + \alpha)\) for each \(\tau_s\) condition.

In addition to the shifts in the resonance-positions of signals, the amplitudes of resonance signals decrease with increasing \(\tau_s\), as seen in Fig. 1(b). If these experimental data are analyzed by using the conventional model without considering a finite-width pulse, \(B_{ac}\) is underestimated and \(f_{ac}\) is overestimated. In contrast, we can evaluate an accurate \(B_{ac}\) value from the experimental data by using our model [Eq. (2)]. Corresponding colored solid lines in Fig. 1(b) are the fitting results according to Eq. (2) with \(f_{ac} = 500\) kHz. These fitting results are consistent with the experimental results, indicating the validity of our proposed model. From these fitting results, we obtained \(B_{ac} = 4.4 \pm 0.1\) \(\mu T\) (red), 4.5 \(\pm 0.1\) \(\mu T\) (blue), and 4.4 \(\pm 0.1\) \(\mu T\) (green) for \(\tau_s = 124\) ns, 374 ns, and 622 ns, respectively. This good agreement of \(B_{ac}\) values indicates the validity of our proposed model. Note that the repetition times among the measurements with various pulse widths were different, and that the various errorbars among the measurements were caused by the difference of the repetition times. Although the errorbars of the measurement with \(\tau_s = 374\) ns were larger than those of the others because of the least repetition times (\(1 \times 10^6\) times), all of the errorbars were sufficiently small to observe the finite-pulse-width effect on AC magnetometry for the asynchronous fields, as shown in Fig. 1(b).

Following the similar procedure of Hahn-echo-based magnetometry, we also investigated AC magnetometry with Carr-Purcell (CP)-type sequences \(^{29,28}\) for AC magnetic fields with randomly distributed \(\phi_{ac}\) values; the averaged magnetometry signal is given by

\[
S_{CP} = J_0 \left[ g_{NV} B_{ac} N \tau (1 + \alpha) W_{N,\tau_s}(\omega_{ac}) \right] e^{-N/T_{2p}(N)^2},
\tag{3}
\]

\[
W_{N,\tau_s}(\omega_{ac}) = \frac{\sin \left( \frac{\omega_{ac} N \tau (1 + \alpha)}{2} \right)}{\omega_{ac} N \tau (1 + \alpha)} \left[ 1 - \frac{\cos \left( \frac{\omega_{ac} \tau}{2} \right)}{\cos \left( \frac{\omega_{ac} (1 + \alpha) \tau}{2} \right)} \right].
\tag{4}
\]

where \(N\) is the number of \(\pi\) pulses and \(T_2(N)\) is the coherence time by using the N-pulse CP sequences. To derive Eqs. (3) and (4), we used phase accumulation due to a synchronous AC field to the CP sequences, represented by \(\Phi_{CP}\) in Ref. 21. As in the case of the Hahn-echo sequence, the Bessel term in Eq. (3) is unity if \(B_{ac} = 0\), and the baseline of CP-based magnetometry signals is thus subjected to exponential decay due to decoherence of spin states in NV centers. We measured the coherence time by using the XY4-1 sequence without the AC field: \(T_2(N = 4) = 124 \pm 5\) \(\mu s\) and \(p = 1.21 \pm 0.08\).

Figure 2 shows the AC magnetometry results using a XY4-1 sequence, \(^{27}\) which is classified as the CP-type sequence and having \(N = 4\) \(\pi\) pulses. The frequency of the AC magnetic field \(f_{ac}\) was set at 500 kHz. The red and blue circles in Fig. 2(b) represent experimental data with \(\tau_s = 126\) ns and 380 ns, which indicate \(\pi\) and \(3\pi\), respectively. The repetition times for the measurements with \(\tau_s = 126\) ns and \(\tau_s = 380\) ns were \(2 \times 10^6\) and \(1 \times 10^6\). Figure 2(b) also shows the calculated result of AC magnetometry using the XY4-1 sequence with \(\tau_s = 0\) (black dashed line). As in the case of Hahn-echo magnetometry, resonance signals for \(\tau_s \neq 0\) (experimental data) shifted toward the smaller value of \(N\) relative to ideal \(\tau = k/2 f_{ac}\) with increasing \(\tau_s\). This result was also understood by the modification of \(\tau\) as \(k f_{ac}(1 + \alpha)\) derived from Eq. (4). In fact, for \(\tau_s = 126\) ns, the two resonance signals were observed around \(N = 3.496\) \(\mu s\) and 11.496 \(\mu s\), which coincide with \(k f_{ac}(1 + \alpha)\) with \(k = 1\) and \(k = 3\), respectively. Corresponding colored solid lines in Fig. 2(b) are the fitting results according to Eq. (4) with

\[\langle \pi/2 \rangle_x, \quad \pi_y, \quad \pi_y, \quad \langle \pi/2 \rangle_x.\]
\( f_{ac} = 500 \) kHz. These fitting results are consistent with the experimental results and yielded \( B_{ac} = 3.7 \pm 0.1 \) \( \mu \)T (red) and 3.6 \( \pm 0.1 \) \( \mu \)T (blue) for \( \tau_x = 126 \) ns and 380 ns, respectively. These results of the CP-type sequences indicate that our model can explain the shift in resonance-signal positions and the decrease in amplitudes of resonance signals with increasing \( \tau_x \), which cannot be predicted by the conventional model without considering a finite pulse width.

Finally, we discuss the signal contrast of AC magnetometry in resonant conditions of the decoupling sequences. Equation (2) under the resonant condition of the Hahn-echo sequences, i.e., \( \tau = k f_{ac} (1 + \alpha) (k = 1, 3, 5, \ldots) \), is represented by

\[
S^R_{\text{Echo}} = I_0 \left[ \frac{2 N V N B_{ac}}{\pi f_{ac}} \cos^2 (\pi f_{ac} \tau_x / 2) \right] e^{-\left(\tau_{\pi T}\right)^2}. \tag{5}
\]

In addition, Eq. (5) under the resonant conditions of the CP-type sequences, i.e., \( \tau = k f_{ac} (1 + \alpha) (k = 1, 3, 5, \ldots) \), is given by

\[
S^R_{\text{CP}} = I_0 N_{\pi N V B_{ac}} \pi f_{ac} \tau_x \left[ N_{\pi f_{ac} \tau_x} \right] e^{-\left(\tau_{\pi T}(N)^2\right)}. \tag{6}
\]

If \( \tau_x = 0, S^R_{\text{Echo}} \propto I_0 \left[ 2 (N_{\pi N V} / 2 \pi) B_{ac} N f_{ac} \right] \), which is consistent with Ref. 18. The Bessel terms in Eqs. (5) and (6) do not depend on \( k \). However, amplitudes of resonance signals decrease exponentially with \( \tau = k f_{ac} (1 + \alpha) \) for the Hahn-echo sequences and \( \tau = k f_{ac} (1 + \alpha) \) for the CP-type sequences, which is given as the exponential terms in Eqs. (5) and (6).

We plot the normalized amplitudes of resonance signals obtained from Figs. (1b) and (2b) as a function of \( f_{ac} \tau_x \) in Fig. 3: blue circles and purple triangles correspond to \( k = 1 \) and 3, respectively, for the Hahn-echo measurements \( (N = 1) \), and red squares and green pentagons correspond to \( k = 1 \) and 3, respectively, for the XY4-1 measurements \( (N = 4) \). Note that \( f_{ac} = 500 \) kHz, \( \tau_x = 124 \) ns, 374 ns, and 622 ns for the Hahn-echo measurements, and \( \tau_x = 126 \) ns and 380 ns for the XY4-1 measurements. Each data was normalized by the photon-count-related term obtained by the fitting to each experimental result. The details of the photon-count-related term were explained in Appendix in Ref. 21. Corresponding colored lines in Fig. 3 are calculations in accordance with Eq. (5) with \( B_{ac} = 4.5 \) \( \mu \)T and Eq. (6) with \( B_{ac} = 3.6 \) \( \mu \)T. As can be seen in Fig. 3, these calculations well reproduce the experimental results, indicating that our proposed model can explain the dependence of AC magnetometry signals on pulse width \( \tau_x \) for asynchronous AC magnetic fields. Note that our model results (solid lines in Fig. 3) implies that the amplitude of a resonance signal decreases with increasing \( f_{ac} \). Therefore, the finite-pulse-width effects should be taken into account for AC magnetometry based on the decoupling sequences unless \( \tau_x \) is considerably shorter than \( 1/f_{ac} \).

In this study, we theoretically and experimentally investigated the finite-pulse-width effect on AC magnetometry using decoupling sequences for asynchronous AC magnetic fields with randomly distributed phase values. As in the case of a synchronous AC field, we revealed that the finite pulse width affects the AC magnetometry spectra of asynchronous AC magnetic fields. For instance, resonance signals appear at lower \( \tau \) values relative to ideal values \( \tau = k f_{ac} (k = 1, 3, 5, \ldots) \) for Hahn-echo-based magnetometry and \( \tau = k f_{ac} / 2 \) for CP-type-sequence-based magnetometry in the precession-time plot. In addition, the amplitudes of resonance signals decrease with increasing pulse width \( \tau_x \). These phenomena cannot be predicted by the conventional model without considering a finite pulse width. Our model also predicted that the amplitudes of resonance signals decrease with increasing \( f_{ac} \). To evaluate accurate amplitude \( B_{ac} \) and frequency \( f_{ac} \) of high-frequency AC magnetic fields from experimental data, therefore, the finite-pulse-width effects should be taken into account unless \( \tau_x \) is sufficiently shorter than \( 1/f_{ac} \).

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**REFERENCES**

1. J. M. Taylor, P. Cappellaro, L. Childress, L. Jiang, D. Budker, P. R. Hemmer, A. Yacoby, R. Walsworth, and M. D. Lukin, *Nat. Phys.* 4, 810 (2008).

2. 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* 455, 648 (2008).

3. L. Rondin, J.-P. Tetienne, S. Rohart, A. Tiwari, T. Higant, P. Spinicelli, J.-F. Roch, and V. Jacques, *Nat. Commun.* 4, 2279 (2013).

4. R. Schirhagl, K. Chang, M. Loretz, and C. L. Degen, *Annu. Rev. Phys. Chem.* 65, 83 (2014).

5. H. J. Mamin, M. Kim, M. H. Sherwood, C. T. Rettnier, K. Ohno, D. D. Awschalom, and D. Rugar, *Science* 339, 557 (2013).

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

7. K. Ohashi, T. Rosskopf, H. Watanabe, M. Loretz, M. Tao, R. Hauert, S. Tomizawa, T. Ishikawa, J. Ishi-Hayase, S. Shikata, C. L. Degen, and K. M. Itoh, *Nano Lett.* 13, 4733 (2013).

8. I. Lovchinsky, J. D. Sanchez-Yamagishi, E. K. Urbach, S. Choi, S. Fang, T. I. Andersen, K. Watanabe, T. Taniguchi, E. Kaxiras, P. Kim, H. Park, and M. D. Lukin, *Science* 355, 503 (2017).

9. N. Aslam, M. Pfender, P. Neumann, R. Reuter, A. Zappe, F. Fávaro de Oliveira, A. Denisenko, H. Sumiya, S. Onoda, J. Isoya, and J. Wrachtrup, *Science* 357, 67 (2017).

10. C. L. Degen, F. Reinhard, and P. Cappellaro, *Rev. Mod. Phys.* 89, 035002 (2017).

11. C. L. Degen, *Nature Nanotech.* 3, 643 (2008).

12. S. Kotler, N. Akerman, Y. Glickman, A. Keselman, and R. Ozeri, *Nature* 473, 61 (2011).

13. 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).
14. L. Viola, E. Knill, and S. Lloyd, Phys. Rev. Lett. 82, 2417 (1999).
15. L. Cywiński, R. M. Lutchyn, C. P. Nave, and S. Das Sarma, Phys. Rev. B 77, 174509 (2008).
16. N. Bar-Gill, L. M. Pham, A. Jarmola, D. Budker, and R. L. Walsworth, Nat. Commun. 4, 1743 (2013).
17. L. M. Pham, N. Bar-Gill, C. Belthangady, D. Le Sage, P. Cappellaro, M. D. Lukin, A. Yacoby, and R. L. Walsworth, Phys. Rev. B 86, 045214 (2012).
18. G. de Lange, D. Ristè, V. V. Dobrovitski, and R. Hanson, Phys. Rev. Lett. 106, 080802 (2011).
19. W. Ma, F. Shi, K. Xu, P. Wang, X. Xu, X. Rong, C. Ju, C.-K. Duan, N. Zhao, and J. Du, Phys. Rev. A 92, 033418 (2015).
20. L. M. Pham, S. J. DeVience, F. Casola, I. Lovchinsky, A. O. Sushkov, E. Bersin, J. Lee, E. Urbach, P. Cappellaro, H. Park, A. Yacoby, M. Lukin, and R. L. Walsworth, Phys. Rev. B 93, 045425 (2016).
21. T. Ishikawa, A. Yoshizawa, Y. Mawatari, H. Watanabe, and S. Kashiwaya, Phys. Rev. Applied 10, 054059 (2018).
22. J. R. Maze, P. L. Stanwix, J. S. Hodges, S. Hong, J. M. Taylor, P. Cappellaro, L. Jiang, M. V. G. Dutó, E. Togan, A. S. Zibrov, A. Yacoby, R. L. Walsworth, and M. D. Lukin, Nature 455, 644 (2008).
23. B. Zhou, P. C. Jerger, K.-H. Lee, M. Fukami, F. Mujid, J. Park, and D. D. Awschalom, arXiv:1903.09287 (2019).
24. D. R. Glenn, D. B. Bucher, J. Lee, M. D. Lukin, H. Park, and R. L. Walsworth, Nature 555, 351 (2018).
25. K. Sasaki, K. M. Itoh, and E. Abe, Phys. Rev. B 98, 121405 (2018).
26. A. Larouari, J. S. Hodges, and C. A. Meriles, Appl. Phys. Lett. 97, 143104 (2010).
27. H. Y. Carr and E. M. Purcell, Phys. Rev. 94, 630 (1954).
28. S. Meiboom and D. Gill, Rev. Sci. Instrum. 29, 688 (1958).
29. T. Gullion, D. B. Baker, and M. S. Conradi, J. Magn. Reson. 89, 479 (1990).