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Temperature-robust diamond magnetometry based on the double-transition method

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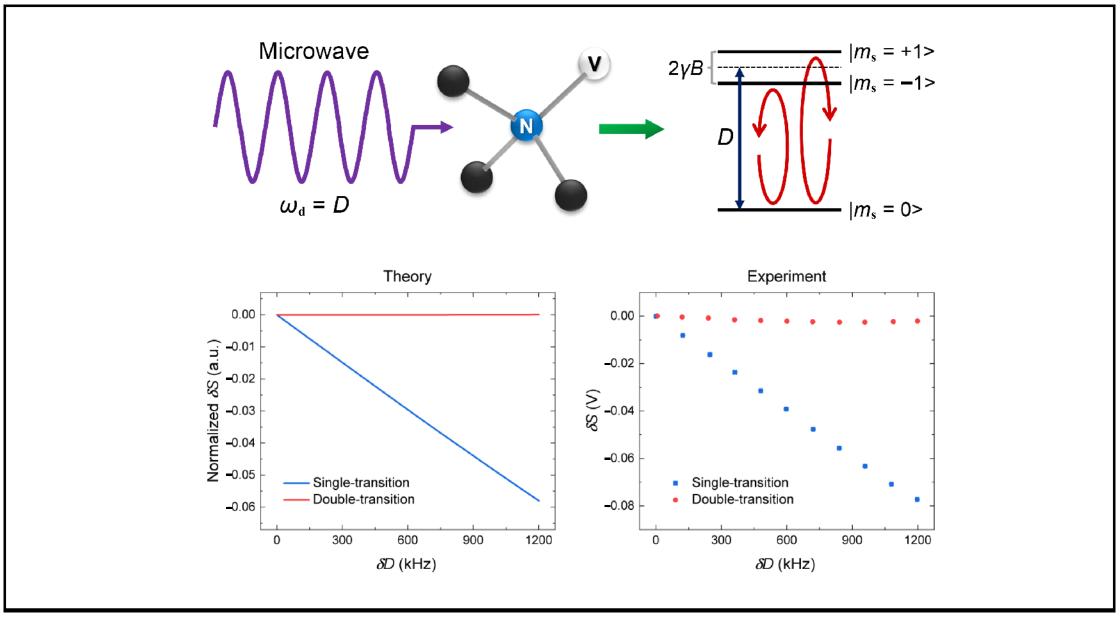
Graphical abstract



Using the double-transition method to resist the impact of temperature drift.

Public summary

- A type of temperature-robust diamond magnetometry based on the double-transition method has been demonstrated, which utilizes both of the transitions between the $|m_s = 0\rangle$ and $|m_s = \pm 1\rangle$ sublevels of the nitrogen-vacancy (NV) electronic ground state with incomplete degeneracy of the $|m_s = \pm 1\rangle$ states.
- With the use of the double-transition method, the variations of fluorescence resulting from the zero-field splitting drifts can be counteracted, and the magnetometry signal drift induced by the temperature drift can be eliminated to the fourth order term in the Taylor expansion, which is safe to be neglected on most occasions.
- The experimental results demonstrate that the magnetometry signal drift in our method has no obvious dependence on the zero-field splitting drift and the drift of magnetic field measurement result have been reduced by about 7-fold, compared with that of the conventional diamond magnetometry only utilizing the transition between $|m_s = 0\rangle$ and $|m_s = +1\rangle$ or $|m_s = -1\rangle$ sublevel.

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Abstract: As a promising solid-state sensor at room temperature, diamond magnetometers based on nitrogen-vacancy (NV) centers have been developed tremendously in recent years. Many studies have demonstrated its potential for achieving high spatial resolution and sensitivity. However, the temperature dependence of the zero-field splitting D of NV centers poses an enormous challenge for the application of diamond magnetometry, since it is difficult to avoid temperature drift in most application scenarios. Here, we demonstrate a type of temperature-robust diamond magnetometry based on the double-transition method. By utilizing both transitions between $|m_s=0\rangle$ and $|m_s=\pm1\rangle$ sublevels with incomplete degeneracy of the $|m_s=\pm1\rangle$states, the impacts of D variations induced by temperature drift can be counteracted. The drift of magnetic field measurement result has been reduced by approximately 7-fold. With further improvements, the temperature-robust diamond magnetometry has the potential to be applied in biomagnetism and space science research.

Keywords: diamond magnetometry; nitrogen-vacancy center; temperature robustness

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1 Introduction

The nitrogen-vacancy (NV) center in diamond is a solid-state spin system with capability of operation under a wide range of conditions[1, 2], which has attracted much interest for being utilized as a quantum magnetometer with nanoscale spatial resolution and potential sensitivity up to femtotesla at room temperature[3]. Due to the different orientations of the NV symmetry axis, it is also possible to use the NV center ensemble to achieve vector magnetometry[4]. The miniature sensing volume of a diamond magnetometer based on NV centers provides an excellent compatibility with the flux concentrators for further sensitivity improvement[5, 6]. The NV diamond magnetometry has the potential to be applied in biomagnetism and geoscience. Previous works have demonstrated the detection of magnetic fields generated by single-neuron action potentials[7] and the magnetic imaging of geological samples[8] using NV centers. These correlational researches have demonstrated the outstanding ability of NV center in magnetic field measurement.

However, due to the temperature-dependent zero-field splitting D of the NV center[9, 10], the NV diamond magnetometry is affected by temperature. For example, in case of that the temperature drift couple to D with $\mathrm{d}D/\mathrm{d}T\approx-74$ kHz/K[9], temperature variation of 1 K will lead to a variation of 74 kHz in the resonance frequency, corresponding to a magnetic field variation of 2.6 μT. Such error is not negligible for application scenarios that need precise measurement of the magnetic field[7, 11]. The common solutions are affixing the diamond to SiC heat sink to stabilize the temperature and compensating feedback to the microwave frequency[4, 7], which can only alleviate the problem to some extent. Previous works also overcame this by monitoring the transitions between $|m_s=0\rangle$ and $|m_s=\pm1\rangle$ sublevels simultaneously[5, 12]. The frequency modulation (FM) of microwaves and strong bias magnetic fields to break the degeneracy of $|m_s=\pm1\rangle$ states absolutely are indispensable for the schemes.

In this article, we demonstrate a type of temperature-robust diamond magnetometry based on the double-transition method. The magnetometry utilizes both of the transitions between $|m_s=0\rangle$ and $|m_s=\pm1\rangle$ sublevels with incomplete degeneracy of the $|m_s=\pm1\rangle$ states, and the impacts of D variations induced by temperature drift can be counteracted. There is no requirement for applying a strong bias magnetic field, which allows the magnetometry to work under low-field condition. The measured drift of the magnetic field measurement result is about 7-fold smaller than of the conventional diamond magnetometry. The temperature-robust diamond magnetometry provides a way to precisely measure the magnetic field under ambient temperature drift, which benefits applications in biomagnetism[7] and space scientific research[13].

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2 Methods

The NV center is a point defect in the diamond, which consists of a substitutional nitrogen adjacent to a vacancy. It allows optical polarization, manipulation, and readout of its spin state[14–16]. The level structure of the NV center is shown in Fig. 1a. The electronic ground state is a spin triplet state $^3A_2$ with the lower sublevel $|m_s=0\rangle$ separated from the $|m_s=\pm1\rangle$ sublevels by a zero-field splitting $D\approx2.87$ GHz at room temperature. The NV center can be optically excited to the excited state ^3E by a 532 nm laser, and decay to the ground state through a spin-conserving transition, which leads to fluorescence emission from 600 nm to 850 nm. The intensity of fluorescence is dependent on the spin state of the NV center. Neglecting the hyperfine coupling, electric field and crystal stress, the Hamiltonian of the ground state with a bias magnetic field $\vec{\boldsymbol{B}}$ is given by Ref. [17]:

$$H=DS_z^2+\gamma\vec{\boldsymbol{B}}\cdot\vec{\boldsymbol{S}}, \tag{1}$$

where $\gamma=28$ GHz/T is the gyromagnetic ratio of the electron spin. $\vec{\boldsymbol{S}}$ is the the dimensionless electronic spin-1 operator. The principle of diamond magnetometry is detecting the Zeeman shift of the ground-state spin levels, which leads to a change in the intensity of spin state-dependent fluorescence.

The temperature-robust diamond magnetometry based on the double-transition method (termed here as double-transition magnetometry) uses the NV center ensemble as the magnetic sensing element for magnetic field measurement. The double-transition method refers to the magnetometry working under an appropriate magnetic field where the degeneracy of $|m_s=\pm1\rangle$ states is not completely broken, and both transitions between $|m_s=0\rangle$ and $|m_s=\pm1\rangle$ sublevels (termed here as $|m_s=0\rangle\leftrightarrow|m_s=\pm1\rangle$ transitions) contribute to the output of magnetometry (termed here as the magnetometry signal). An amplitude-modulated microwave with frequency $\omega_d=D$ is applied to transfer the population into both $|m_s=\pm1\rangle$ sublevels and modulate the fluorescence into the high frequency domain to overcome the flicker noise. The modulated fluorescence is converted to a photocurrent and sent into a lock-in amplifier (LIA) for demodulation. The demodulated signal serves as the magnetometry signal.

Considering the temperature drift that leads to a small variation of zero-field splitting δD, which will cause the resonance frequencies of $|m_s=0\rangle\leftrightarrow|m_s=\pm1\rangle$ transitions to change identically, the impact of δD can be equivalent to the variation of ω_d. Due to the inverse slopes of optically detected magnetic resonance (ODMR) spectra corresponding to $|m_s=0\rangle\leftrightarrow|m_s=\pm1\rangle$ transitions at $\omega_d=D$, the changes in fluorescence induced by δD will be counteracted. Hence, the magnetometry can resist the impact of temperature drift. Conversely, for the small variation in magnetic field δB that causes the resonance frequencies of $|m_s=0\rangle\leftrightarrow|m_s=\pm1\rangle$ transitions to change inversely, the variation in magnetometry signal δS has a linear relationship with δB, which means that the system is sensitive to the magnetic field.

A schematic of the experimental setup is shown in Fig. 1b, including laser excitation, microwave generation, and magnetic field detection. The diamond is cut into a piece with a size of 1.2 mm × 1.2 mm × 0.4 mm, and the {110} facet (1.2 mm × 1.2 mm facet) is perpendicular to the bias magnetic field applied by a coil. The two aluminum slices are used to clamp the diamond, and the two resistors heated by the RF

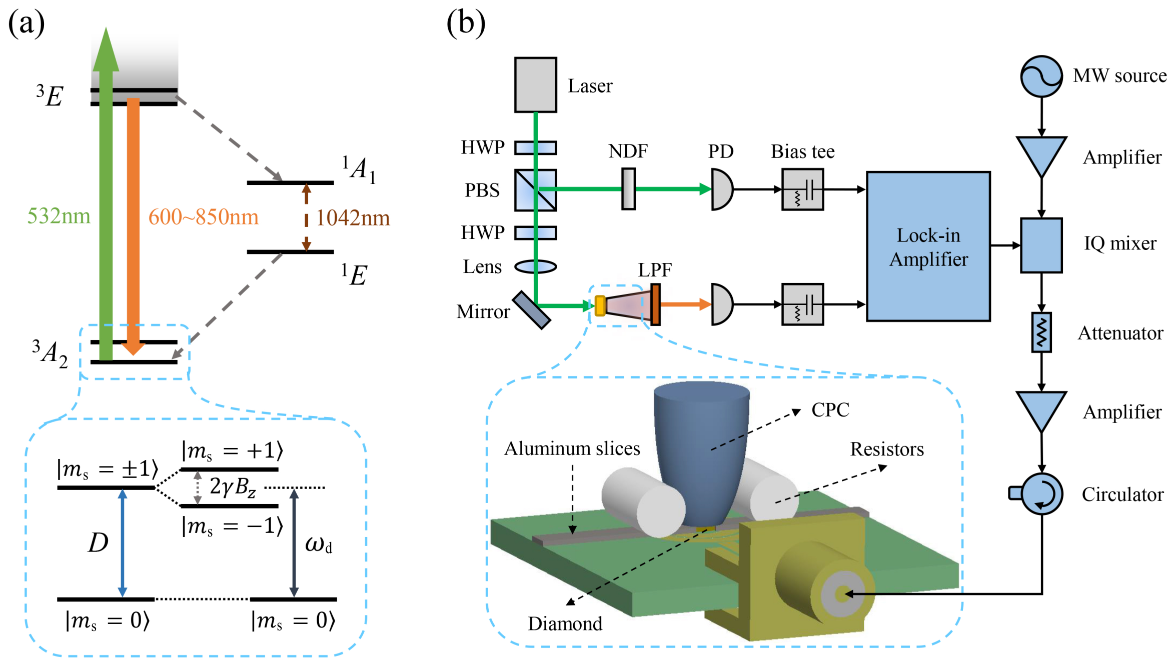

Fig. 1. The level structure of NV center in diamond and the setup of temperature-robust diamond magnetometry based on the double-transition method. (a) Energy-level diagram for the NV center. The external magnetic field B_z applied along the NV symmetry axis lifts the degeneracy of $|m_s=\pm1\rangle$ sublevels with a Zeeman shift. (b) The schematic of experimental setup. The green and red arrows represent for the 532 nm laser and fluorescence. A few of components used in the experimental setup like the coil for applying bias magnetic field are not shown in the figure.

signal with a frequency of 80 MHz are used to change the temperature of the diamond. The 532 nm laser is used to optically pump NV centers. A compound parabolic concentrator (CPC) is clung to the diamond for collecting fluorescence. After being filtered by a 650 nm longpass filter (LPF), the fluorescence is transferred to a photodetector (PD). A portion of the 532 nm laser separated by the polarizing beamsplitter (PBS), as the reference light, is transferred to another PD for cancellation of laser noise[7, 18]. The intensity of the reference light is adjusted by a neutral density filter (NDF) and a half-wave plate (HWP) to optimize noise cancellation. Another HWP is used to adjust the laser-polarization direction. Both of the PDs are connected to the bias tees. The DC outputs of bias tees are monitored by an oscilloscope, and the RF outputs are sent into an LIA. A microwave (MW) source is used to generate a microwave at frequency $\omega_{\rm d}$, which is transmitted to the in-phase/quadrature (IQ) mixer and multiplied by a modulation signal from the LIA for amplitude modulation (AM). Since the output power of the MW source is not enough to reach the input threshold of IQ mixer, an amplifier is added into the circuit between the MW source and the IQ mixer. The amplitude-modulated microwave is amplified by another amplifier and sent to a double split ring resonator (DSRR)[19] to manipulate the spin state of the NV center. The attenuator and circulator are used to protect the amplifier.

We presented a comparison between the double-transition magnetometry and the conventional diamond magnetometry based on frequency modulated microwave[6, 7, 18]. The diamond magnetometry based on frequency modulated microwave, which only utilizes the $|m_{\rm s}=0\rangle \leftrightarrow |m_{\rm s}=+1\rangle$ or $|m_{\rm s}=0\rangle \leftrightarrow |m_{\rm s}=-1\rangle$ transition, is called single-transition magnetometry in this article. Since the temperature drift will change the resonance frequencies, the single-transition magnetometry suffers from the impact of temperature drift.

Based on the Lorentzian profile of the ODMR spectrum[7, 20], for the two types of magnetometry, the relationship between the zero-field splitting drift δD and magnetometry signal drifts δS induced by it can be calculated as (see Section II of Supporting information)

$$\delta S_{\rm ST} \approx -\frac{3\sqrt{3}}{4}\frac{U_{\rm ST}F_0L_0C}{\Gamma}\delta D, \tag{2}$$

$$\delta S_{\rm DT} \approx \frac{27}{4}\frac{U_{\rm DT}F_0L_0C}{\Gamma^4}(\delta D)^4, \tag{3}$$

where $S_{\rm ST}$ and $S_{\rm DT}$ are the magnetometry signal drifts of single-transition and double-transition magnetometry. $U_{\rm ST}$ and $U_{\rm DT}$ are the dimensionless prefactors introduced by the modulation-demodulation protocol for the single-transition and double-transition magnetometry. Their values are determined by the modulated fluorescence. F_0 is the intensity of fluorescence under nonresonance condition. L_0 is the conversion coefficient between the fluorescence and magnetometry signals in voltage units. Its value is determined by the settings of the LIA. C is the contrast of the ODMR spectrum. Γ is the full width at half maximum (FWHM) of the ODMR spectrum.

The simulation results of the relationships between δS and δD for the two types of magnetometry are shown in Fig. 2.

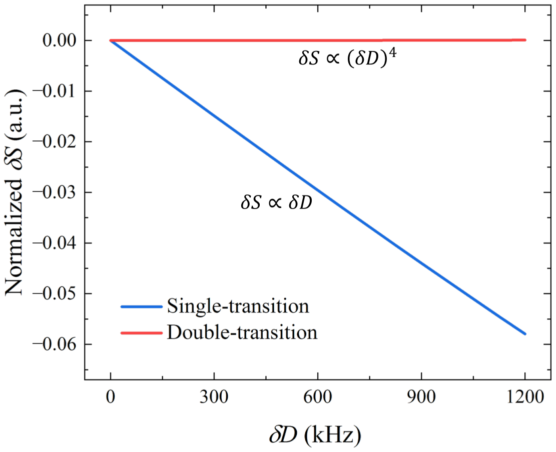

Fig. 2. The simulation results of magnetometry signal drifts δS as the function of zero-field splitting variation δD for the single-transition and double-transition magnetometry. For the double-transition magnetometry, the normalized δS is immune to δD.

The δS was normalized by setting $F_0 = L_0 = C = 1$. The Γ used in the simulation was set as 15 MHz, which is a typical value for the ODMR spectrum with laser and microwave power broadening[20]. Based on the relationship $\mathrm{d}D/\mathrm{d}T \approx -74$ kHz/K, the maximum $\delta D = 1200$ kHz in the simulation corresponds to a temperature variation over 16 K. The magnetometry signal drift of double-transition magnetometry under such a temperature drift is much smaller than that of the single-transition magnetometry.

3 Results and discussion

To experimentally demonstrate the effectiveness of the double-transition method, we measured the magnetometry signal drift δS with different variations of zero-field splitting δD and compared it with that of the single-transition magnetometry. All measurements were carried out with the same diamond in a magnetic shield. The bias magnetic fields applied by a coil in the magnetic shield were 304 μT and 194 μT for the single-transition and double-transition magnetometry. The direction of the bias magnetic field is perpendicular to the {110} facet of diamond. The parameters in the experimental setup, such as the laser and microwave power, were optimized before the measurements. The δD was changed by varying the temperature of the diamond with the resistors. The experimental results are shown in Fig. 3. For the single-transition magnetometry, δS changes with δD in a linear relationship which conforms expectations. Conversely, for the double-transition magnetometry, δS is much smaller than that of the single-transition magnetometry and shows no obvious dependence on δD. With the use of the double-transition method, the influence of temperature drift on diamond magnetometry can be suppressed.

The output of double-transition magnetometry still has slight drift during the measurement. Several factors are reasonable. The first is the temperature dependence of the photoluminescence intensity of NV centers[21, 22] which introduces extra temperature-dependent signal drift. In addition, the thermal expansion of the material caused by temperature drift

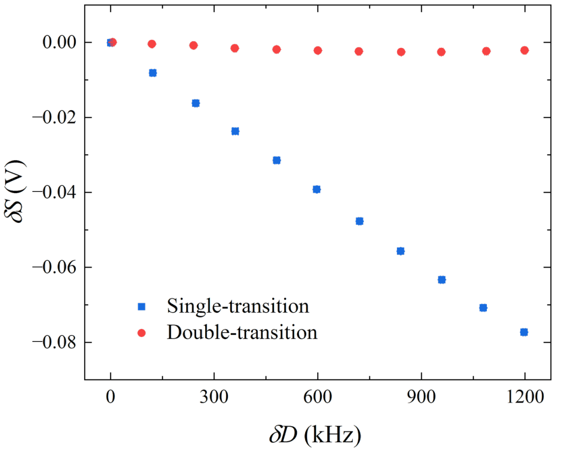

Fig. 3. The measured magnetometry signal drifts δS as the function of zero-field splitting variation δD for the single-transition and double-transition magnetometry. Each data point was acquired for 5 s and averaged. The error bars are smaller than the data points.

will change the mechanical structure and lead to a shift in the laser's excited area in the diamond, which also contributes to the magnetometry signal drift. Noise from other sources such as the fluctuation of laser output power, will also increase the δS of double-transition magnetometry.

To demonstrate the temperature robustness of double-transition magnetometry under working conditions, the magnetometry signal drift δS with temperature drifts δT of the single-transition and double-transition magnetometry were measured. The time domain signals of the two types of magnetometry were acquired for approximately 6000 s. Each data point was recorded for 1 s and averaged. In the first half of the signal acquisition process, the diamond was heated using the resistors. The temperature of the diamond is monitored by a temperature sensor. The δS was converted to the drift of the magnetic field measurement result δB_{mea} using the maximum slope $|\mathrm{d}S/\mathrm{d}B|$ in the linear region of the corresponding method. Fig. 4 shows the δT and δB_{mea} varying with the measuring time in the two methods. Since the compensation feedback of heating power is not supported by our heating equipment, the relationships between temperature and measuring time of the two methods have slight differences.

As shown in Fig. 4a, for the single-transition magnetometry, δB_{mea} has a significant temperature dependence. Conversely, based on the experimental data shown in Fig. 4b, δB_{mea} is almost immune to δT in the double-transition magnetometry. The maximum δB_{mea} (absolute value) in double-transition magnetometry is about 7-fold smaller than that in single-transition magnetometry. The results demonstrate the temperature robustness of our method under working conditions, which allows magnetometry to work under significant ambient temperature drift, such as in the outer space[23, 24]. Since the frequency of temperature drift is typically less than 100 Hz[6], the temperature robustness of magnetometry can also improve the long-term stability of the system, which is important for precision magnetometry applications that require long acquisition times[25].

It has been discussed before that the instability of the mechanical structure and the fluctuation of laser output power can increase the δB_{mea} in double-transition magnetometry. For further improvement, the mechanical structure should be carefully designed to compensate for the thermal expansion of the material and fabricated by materials with a low coefficient of thermal expansion. Additionally, the fluctuation of laser output power should be reduced by using a laser with higher stability. With these improvements, δB_{mea} can be further reduced for the double-transition magnetometry.

Following the method of previous works[5, 6, 26], we measured the maximum slope $|\mathrm{d}S/\mathrm{d}B|$ of double-transition magnetometry as (636 ± 1) V/T and demonstrated the noise floor as $(0.27\pm0.06)\ \mathrm{nT}/\sqrt{\mathrm{Hz}}$ with the microwave frequency being set far from the zero-field splitting D. The major factor that leads to the low $|\mathrm{d}S/\mathrm{d}B|$ is the nonsinusoidal waveform of modulated fluorescence. Due to the noise from the IQ mixer used for AM[27, 28] and other sources such as the low-frequency vibration of meachanical structure, the magnetic amplitude spectral density (ASD) that demondstrates the

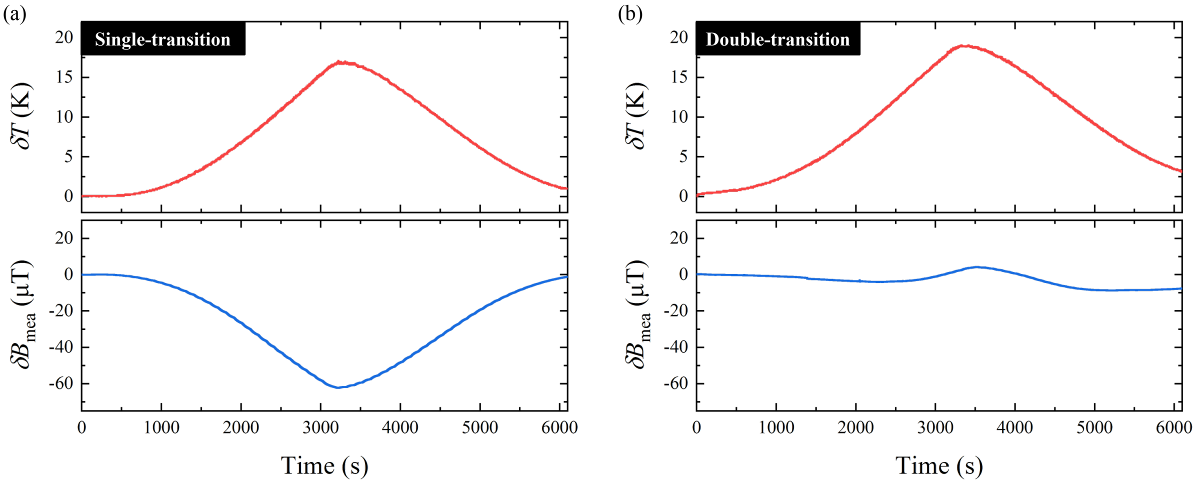

Fig. 4. Comparison between the single-transition and double-transition magnetometry working under significant temperature drifts. (a), (b) Time domain temperature drifts δT and magnetic field measurement result drifts δB_{mea} of the two types of magnetometry.

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sensitivity is $1.6\ \mathrm{nT}/\sqrt{\mathrm{Hz}}$ at 10 Hz (see Section III of Supporting information). For further improvement, a better modulation-demodulation protocol should be established.

The double-transition method is different from the dual-resonance modulation technique used in previous work[5]. In the double-transition method, both of the $|m_s = 0\rangle \leftrightarrow |m_s = \pm 1\rangle$ transitions are utilized with incomplete degeneracy of the $|m_s = \pm 1\rangle$ states. This allows the magnetometry based on it to work under low-field conditions.

4 Conclusions

In summary, we have demonstrated a type of temperature-robust diamond magnetometry based on the double-transition method. By utilizing both of the $|m_s = 0\rangle \leftrightarrow |m_s = \pm 1\rangle$ transitions with incomplete degeneracy of the $|m_s = \pm 1\rangle$ states, the variations of fluorescence resulting from zero-field splitting drifts can be counteracted. The measured drift of the magnetic field measurement result is about 7-fold smaller than that of the single-transition magnetometry. Temperature robustness allows the magnetometry to work under significant ambient temperature drift and improves the long-term stability of the system. The magnetometry can be further improved by optimizing the modulation-demodulation protocol and combining it with the sensitivity enhancing techniques, such as light-trapping diamond waveguide[18], surface coating[29], and ^{12}C isotopic enrichment[30], to achieve high sensitivity in the future. The improved temperature-robust diamond magnetometry has the potential to be applied in biomagnetism and space scientific research.

Supporting information

The supporting information for this article can be found online at https://doi.org/10.52396/JUSTC-2022-0150. The supporting information includes the experimental setup, the principle of double-transition magnetometry, and the further improvements. There are two figures and one table.

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Conflict of interest

The authors declare that they have no conflict of interest.

Biographies

Caijin Xie is a postgraudate student of University of Science and Technology of China. His research focuses on quantum sensing.

Xing Rong is a Professor at the University of Science and Technology of China (USTC). He received the B.S. degree in 2005 and the Ph.D. degree in 2011 from USTC. He joined the Department of Modern Physics of USTC as a Professor in 2014. His research interest includes quantum control of spins in solids, quantum computation, quantum sensing, and exploring new physics beyond the standard model with micrometer scale.

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