Ultra-sensitive hybrid diamond nanothermometer

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Abstract

Nitrogen-vacancy (NV) centers in diamond are promising quantum sensors for their long spin coherence time under ambient conditions. However, their spin resonances are relatively insensitive to non-magnetic parameters such as temperature. A magnetic-nanoparticle-nanodiamond hybrid thermometer, where the temperature change is converted to the magnetic field variation near the Curie temperature, was demonstrated to have enhanced temperature sensitivity (11 mK Hz$^{-1/2}$) [Phys. Rev. X 8, 011042 (2018)], but the sensitivity was limited by the large spectral broadening of ensemble spins in nanodiamonds. To overcome this limitation, here we show an improved design of a hybrid nanothermometer using a single NV center in a diamond nanopillar coupled with a single magnetic nanoparticle of copper-nickel alloy, and demonstrate a temperature sensitivity of 76 μK Hz$^{-1/2}$. This hybrid design enables detection of 2 millikelvin temperature changes with temporal resolution of 5 milliseconds. The ultra-sensitive nanothermometer offers a new tool to investigate thermal processes in nanoscale systems.

Keywords:

Nano-thermometry, Diamond, Nitrogen-vacancy center, Quantum sensing, Magnetic nanoparticle
Introduction

Nanoscale temperature measurement with high sensitivity is important to investigating many phenomena such as thermal mapping of nano-/micro-electronics [1], thermoplasmonics of nanoparticles [2], chemical reactions in nanoliter volume [3], and thermal processes in live systems [4–6]. To probe the thermal dynamics on the nanoscale, various measurement protocols have been developed. The optical thermometers convert the local temperature variation to changes of optical lifetimes [7], fluorescence intensities [8], Raman shifts [9], or emission spectra [10]. Being a non-contact and convenient method, optical-based nanothermometers, like fluorescence proteins [11], dyes [7], and rare-earth nanoparticles [12], have been proposed and demonstrated for temperature detection under various conditions. However, this method has a relatively low sensitivity (typically $1 \text{ K Hz}^{-\frac{1}{2}}$) [13–15] and moreover some optical sensors are subjected to artifacts induced by the local environments of the sensors such as refractive indices and pH values [16]. Electronic temperature measurements, such as scanning thermal microscopy and superconducting quantum interference devices (SQUID), have high spatial resolution and high sensitivity ($\sim 1 \mu\text{K Hz}^{-\frac{1}{2}}$) [17,18], but they require extreme operating conditions and are subjected to contact-related artifacts.

The recent development of diamond-based thermometers provides a promising alternative [19–21]. Nitrogen-vacancy (NV) centers in diamond have long spin coherence time under ambient conditions [22]. Their spin resonance frequencies shift with the environmental temperature [23], which is robust against artifacts from local environments. With photo-stability of NV centers [24], high thermal conductivity [25] and bio-compatibility of the diamond material [26,27], diamond-based thermometers are a potential candidate for temperature sensing in complex systems without the requirement of extreme operating conditions. However, the temperature dependence of NV center spin transition frequencies ($dD/dT \approx -74 \text{ kHz K}^{-1}$) is relatively small. Thus there arises the idea of hybrid diamond thermometers [28,29], in which the temperature change is transduced to a magnetic signal to be detected by the NV center spins. A hybrid nanothermometer composed of a single copper-nickel alloy magnetic nanoparticle (MNP) and a diamond nanocrystal with ensemble NV centers [28] was demonstrated to have a sensitivity as high as $11 \text{ mK Hz}^{-\frac{1}{2}}$ near the Curie temperature of the magnetic nanoparticle, where a small temperature change leads to a large magnetic field change due to the critical magnetization. However, the sensitivity of this hybrid nanothermometer was limited by the short coherence time of ensemble NV centers in nanodiamonds as well as the ODMR linewidth broadening due to the large gradient of the magnetic field from the magnetic nanoparticle. To overcome this limitation, here we constructed a hybrid nanothermometer employing a single NV center in a diamond nanopillar and a single copper-nickel alloy nanoparticle. This design has the following advantages: the spin coherence time of the single NV center is much longer than those in nanodiamonds, and the field gradient induced broadening of ensemble NV centers in nanodiamond is eliminated [28]. Although the photon count rate of a single NV center is lower than those of ensemble NV centers in nanodiamonds, the pillar waveguide configuration largely enhances the photon collection efficiency [30]. We constructed the hybrid nanothermometer by placing the magnetic nanoparticle close to the diamond nanopillar via nano-manipulation based-on atomic force microscopy (AFM). Such a hybrid nanothermometer has a temperature...
sensitivity of 76 μK Hz\(^{-1/2}\), enabling detection of 2 millikelvin temperature changes with temporal resolution of 5 milliseconds. To the best of our knowledge, this is the most sensitive nanothermometer working under ambient conditions. Employing this hybrid sensor, we monitored the temperature changes of a laser heating process and environment temperature fluctuations, as well as thermal dissipation near the sensor when additional heating to the system was induced by controlling the current passing through the microwave antenna. This ultra-sensitive hybrid nanothermometer offers the opportunities of studying fast thermal processes in nanostructures and/or in living systems.

**Results**

The hybrid diamond nanothermometer is composed of a single NV center in a diamond pillar and a copper-nickel alloy MNP, as illustrated in the inset of Fig. 1a. The ground state of an NV center is a spin triplet. The simplified spin Hamiltonian can be written as

\[
H = D S^2 + \gamma_e B \cdot S,
\]

where \(S\) is the spin operator, \(B\) is the external magnetic field, \(D \approx 2.87\) GHz is the zero-field splitting between the \(m_s = 0\) and \(m_s = \pm 1\) states, and the electron gyromagnetic ratio \(\gamma_e = 2.8\) MHz Gauss\(^{-1}\). The transition frequencies between different spin states can be measured by optically detected magnetic resonance (ODMR) spectroscopy using the spin-dependent fluorescence and resonant microwave manipulation of the spin. Unlike conventional diamond thermometry based on the temperature dependence of \(D\), which has a susceptibility of \(dD/dT \approx -74\) kHz K\(^{-1}\), the hybrid nanothermometer measures the magnetization change of the MNP induced by temperature variation [28]. Near the critical point of the MNP, the temperature susceptibility is large and hence a high temperature sensitivity can be achieved. Figure 1a shows a simulated demagnetization curve when an MNP undergoes the ferromagnetic-paramagnetic transition under a small external magnetic field (100 Gauss). The magnetization of the MNP changes drastically when the temperature approaches the Curie point \((T_C)\). The magnetic field from the MNP induces the Zeeman splitting between the \(m_s = -1\) and \(m_s = +1\) states of the NV center, which can be measured through the ODMR spectroscopy. Using a single NV center in a diamond nanopillar has several advantages over the previous hybrid configuration [28]. First, single NV centers in diamond have longer coherence times than ensemble NV centers in nanodiamonds. In our experiments, the selected NV center has a dephasing time \(T_2^* \approx 1.5\) μs (see Supplementary Figure 2 for details of optical and spin properties of the NV center). As a comparison, the typical dephasing time of NV centers in nanodiamonds is \(\sim 100\) ns [31]. Second, although the fluorescence intensity of a single NV center is lower than that of ensemble NV centers in nanodiamonds, the waveguide effect of the nanopillar structure makes the emission more directional and therefore enhances the fluorescence collection efficiency [30]. Third, the large inhomogeneous broadening of the ODMR of ensemble NV centers in nanodiamonds due to the gradient of the magnetic field from MNPs is absent in the case of single NV centers.
The hybrid nanothermometer was constructed by nanomanipulation in an AFM setup. A key factor of the hybrid nanothermometer is the effective coupling between the MNP and the NV center, which strongly depends on their distance and relative orientation. Figure 1b shows an example of nanomanipulation. A much larger splitting of the $m_s = \pm 1$ states appeared in the ODMR spectra when the MNP was pushed closer to the diamond nanopillar. Apart from the coupling strength between the MNP and the NV center, the working range of the hybrid sensor is tens of Kelvin below the Curie temperature. The Curie temperature can be designed by tuning the chemical composition of the copper-nickel alloy nanoparticle. Thus, the hybrid sensor can have a broad working range from cryogenic temperatures to about 600 K [28].

Figure 1. Design of a hybrid nanothermometer composed of a single magnetic copper-nickel alloy nanoparticle and a single nitrogen-vacancy (NV) center in a diamond nanopillar. a, Simulation of the magnetization $M$ of a copper-nickel alloy nanoparticle as a function of temperature under a magnetic field of 100 Gauss. The inset illustrates the configuration of the hybrid nanothermometer. b, Atomic force microscopy (AFM) image of the copper-nickel alloy magnetic nanoparticle (MNP) and the diamond nanopillar before the nanomanipulation (upper graph) and after the nanomanipulation (lower graph), and the corresponding optically detected magnetic resonance (ODMR) spectra of the single NV center before and after nanomanipulation (dots being measurement data and lines the double Lorentzian peak fitting). Scale bar is 1 μm. c, ODMR spectra of the hybrid nanothermometer at different environment temperatures (from 298 K to 324 K from bottom to top). d, ODMR frequency shifts in the heating (blue) and cooling (red) processes. The inset shows the temperature susceptibility of the hybrid nanothermometer, which has the maximum $df/dT \sim 47$ MHz K$^{-1}$ (at 311 K).
To characterize the temperature response of the magnetization of the copper-nickel alloy MNP, we measured the magnetic field at the NV center using continuous-wave ODMR spectroscopy. The environment temperature was controlled by a ceramic heater and calibrated by monitoring the $D$ shift of a reference NV center that is far from any MNP (therefore under zero magnetic field) (see Supplementary Figure 3). After the temperature calibration, a magnetic field of 192 Gauss was applied to enhance the local magnetic field generated by the MNP. Figure 1c plots a series of ODMR spectra of the hybrid nanothermometer at different temperatures. The resonance dips indicate the transition between the $m_s = 0$ and $m_s = -1$ states of the NV center spin. The spin resonance frequencies at different temperatures are plotted in Fig. 1d. With increasing the environment temperature, the resonance frequency splitting was reduced due to the thermal demagnetization of the MNP. The magnetization of the MNP presented a sensitive response. The inset in Fig. 1d summarizes the temperature susceptibility $df/dT$ of the NV center spin resonance frequency ($m_s = -1$ state). At 38 °C, the susceptibility reached its maximum of 47 MHz K$^{-1}$. Comparing to the temperature dependent $D$ shift of an NV center spin, $dD/dT \approx -74$ kHz K$^{-1}$, the temperature susceptibility of the hybrid nanothermometer is enhanced approximately 600-fold. Furthermore, the magnetization and demagnetization of this MNP is reversible under the external magnetic field (192 Gauss) alignment, as evidenced.
by the overlap between the temperature responses during the heating and cooling processes (Fig. 1d). The reversibility and chemical stability of the hybrid nanothermometer were verified by repeating more heating/cooling measurements on the same hybrid sensor at different times (See Supplementary Figure 4).

For high-precision temperature measurement, it is important to exclude the laser heating effect. In conventional optical-based nanothermometers, laser heating on the thermometers induces a local temperature increase [32] and then the measurement of environmental temperature is complicated by the laser heating effect. Laser heating also exists in our hybrid nanothermometer. However, the pulsed ODMR protocol allows to largely reduce the laser heating effect since the laser can be turned off during the spin evolution period in pulsed measurement. The protocol of the pulsed measurement is as follows (see inset of Fig. 2a): First a laser pulse was applied to initialize the spin; then after a waiting time $t_w$ a microwave was applied; and finally a laser pulse was applied after time $t_r - t_w$ to read out the spin state (which also serves to initialize the spin for the next shot of measurement). The interval ($t_r$) between laser pulses was kept constant to keep the heating effect the same for various waiting time ($t_w$) taken between the laser and microwave pulses. To understand the cooling dynamics in the hybrid nanothermometer, we carried out pulsed ODMR measurement with different waiting times $t_w$. The environment temperature was set at 38 °C where the temperature susceptibility attains the maximum $df/dT = 47 \text{ MHz K}^{-1}$. Considering the environment temperature would change with millikelvin scale during the long-term measurement, a reference ODMR measurement with waiting time $t_w = 10 \mu s$ between the laser and microwave pulses was performed simultaneously to calibrate the spin resonance frequency drift due to long term temperature fluctuation. Figure 2a shows the temperature dynamics of the hybrid sensor as a function of the waiting time $t_w$. It shows that the pulsed laser excitation (300 ns) of the hybrid nanothermometer induced a local temperature increase of about 20 mK. Such an increase of temperature is several times larger than temperature fluctuations of interest in, e.g., nanoelectronics and biological systems [4,17]. After the laser was turned off, the local temperature decayed exponentially and recovered to the environment temperature within a time scale of $\sim 1.5 \mu s$. The laser heating effect can be largely reduced by choosing a waiting time $t_w \geq 1.5 \mu s$. In the following experiments, we chose $t_w = 1.5 \mu s$ to reduce the laser heating effect while still having a reasonable measurement duty ratio.
To determine the temperature sensitivity of the hybrid nanothermometer, free-induction decay (FID) of the NV electron spin was measured. The result is plotted in Fig. 2b. The pulse sequence of the FID measurement (inset of Fig 2b) was modified to reduce the laser heating effect ($t_w = 1.5 \mu$s) while the interval between the laser pulses was kept constant so that the total laser power applied to the sample was the same for different FID time (see Supplementary Figure 5 for comparison of FID signal with and without the pulse modification). At the maximum temperature susceptibility point (38 °C), the sensitivity of the hybrid nanothermometer is estimated to be 76 $\mu$K Hz$^{-1/2}$ (see Methods for details of the sensitivity estimation). Consistent results were obtained from two other hybrid sensors, revealing the robustness and reproducibility of our hybrid quantum thermometer design (see Supplementary Figure 6). To further verify that the sensitivity was shot-noise limited, we carried out real-time FID measurement with an optimized waiting time of 983 ns (where we had the maximum resonance frequency susceptibility of the FID signal, see Methods for details). The linear dependence of the temperature accuracy (defined as the standard derivation of the temperature measurements $\sigma_T$) on the inverse square root of integration time (see Fig. 2c) indicates that the sensitivity is shot-noise limited; with a shot-noise limited sensitivity in the real-time measurement of about 87 $\mu$K Hz$^{-1/2}$. With such high sensitivity, our hybrid nanothermometer provides the capability of measuring millikelvin temperature dynamics with a temporal resolution of a millisecond. For example, Fig. 2d illustrates the histogram of the uncertainty of the measured temperatures with a sampling time of 5 ms. The distribution presents Gaussian statistics with a standard deviation of 1.5 mK. As a comparison, the previous version of nanodiamond-based hybrid sensor (with sensitivity of 11 mK Hz$^{-1/2}$) [28] would need 50 seconds of measurement to achieve the same precision. Nearly two orders of magnitude enhancement of the sensitivity thus enables a wide range of applications, especially in measuring millikelvin temperature change (induced by environmental fluctuation, laser heating, or dissipation from micro/nanostructures) with high temporal resolution. To demonstrate the hybrid nanothermometer as a powerful temperature

Figure 3. Real-time monitoring of local thermal dynamics. a to c, Environment temperature fluctuation measured by the hybrid nanothermometer with various data integration times (0.5 s, 40 ms and 10 ms, in a, b, and c, respectively).
monitor, we performed environment temperature tracking. The environment temperature dynamics at various timescales was measured and shown in Figs. 3a to c. The temperature fluctuation has a maximum amplitude of $\pm 10 \text{ mK}$, $\pm 5 \text{ mK}$, and $\pm 2 \text{ mK}$, at timescales of 100, 1, and 0.1 second, respectively.

The hybrid nanothermometer has potential applications in monitoring thermal dynamics in microscopic systems such as biological thermal processes and heat dissipation in micro-/nano- electronic devices. For a proof-of-the-principle experiment, we utilized the microwave antenna around the hybrid nanothermometer as a heating source. We coupled a chopped DC current into the microwave stripline (which has a width of 20 μm and is located ~25 μm away from the sensor) using an RF/DC combiner. The heat generation/dissipation dynamics was monitored by real-time tracking of the local temperature at the location of hybrid nanothermometer. The chopped DC current was chopped, an instantaneous change in the magnetic field induced by the DC current resulted in a sudden jump of the spin resonance frequencies in the ODMR, while heating and cooling processes were observed as evidenced by the subsequent spin resonance frequency shift after the DC current chopping. The temperature variation is plotted in the lower panel of Fig 4a, in which the $\Delta T = 0$ point is defined by the average of the data at the steady state of the heating/cooling process.

Figure 4. Heat dissipation dynamics in the hybrid nanothermometer under pulsed heating. a, Upper figure shows the chopped DC current passing through the microwave stripline. Middle figure plots the corresponding ODMR spectra of the NV center in the hybrid nanothermometer. The sudden shift of the ODMR frequency is due to the magnetic field from the chopped DC current. The lower figure is the temperature variation of the hybrid sensor under heating by the chopped current. b, The heating and cooling dynamics measured by the hybrid nanothermometer. The $\Delta T = 0$ point is defined by the average of the data at the steady state of the heating/cooling process.

The observation about the thermal dynamics was verified by a control experiment where alternating current with...
forward and reverse directions was applied with constant heating power. No frequency shift was observed following the jump caused by the electric magnetic fields (see Supplementary Figure 7 for more details). This demonstration experiment proves the potential of the hybrid nanothermometer as a diagnostic tool for studying the thermal dissipation in microelectronics with high spatial and temperature resolution.

Conclusion and discussion

In conclusion, we developed an ultra-sensitive hybrid nanothermometer composed of a single NV center in a diamond nanopillar and a magnetic nanoparticle. When the environment temperature changed near the critical temperature of the MNP, the magnetic field generated by the MNP abruptly changed. The magnetic field change was readily measured by the ODMR of the NV center. The sensitivity of the hybrid nanothermometer is as high as 76 μK Hz$^{-1/2}$. The high temperature sensitivity indicates fast data acquisition yet with a high temperature measurement precision. We applied the sensor to monitor the environment fluctuations as well as the in-situ heat dissipation dynamics. Stable environment temperature and large dynamic range are critical for further exploration of our hybrid nanothermometer to measure small temperature variation in systems of interest. In fact, the dynamic range of the hybrid sensor can be further enhanced by the frequency-locking scheme for the NV magnetometry [33].

To further improve the sensitivity of the hybrid sensor, we can increase the temperature susceptibility of the NV center resonance frequency. A 4-fold enhancement of the susceptibility is predicted theoretically by using a 200 nm diameter MNP and locating an NV center 25 nm from the MNP [28]. In addition, longer coherence time $T_2^*$ will improve the sensitivity as $\sim 1/\sqrt{T_2^*}$ [34]. The dephasing time $T_2^*$ can be increased to 90 μs by using an NV center in isotopically purified diamond [22,35], which means a 7-fold improvement of sensitivity. The performance of the nanosensors depends on their specific configuration and therefore each nanosensor needs to be characterized individually before measurement is carried out in realistic systems. To make the performance of the hybrid sensors (see Fig.1 and Supplementary Figure 6) more reproducible, one can employ MNPs with uniform size and composition [36] and control the MNP proximity to the NV center with AFM manipulation. Another method to stabilize the performance is to coat a magnetic alloy thin film (by, e.g., thermal evaporation deposition) on diamond pillars with controllable thickness and composition. A further improvement of the hybrid nano-thermometer design is to replace the diamond nanopillar with a diamond cantilever [37,38] so that a scanning nano-thermometer can be realized with high spatial resolution. Such a design would allow one to extend the present prototypical study of the heat dynamics induced by currents in microwave striplines (Fig. 4) to applications in realistic microelectric devices. An array of the hybrid nano-thermometers may also be constructed to measure the spatial distribution of temperature.

The ultra-sensitive hybrid nanothermometer is especially useful in measuring millikelvin temperature variation with high temporal resolution. As compared with existing ultrasensitive nano-thermometers (such as the SQUID-based nano-thermometer [17,18]), the hybrid diamond nano-thermometer features the applicability in ambient conditions. The new sensor may facilitate the study of a broad range of thermal processes, such as nanoscale chemical reactions, nano-plasmonics, heat dissipation in nano-/micro-electronics, and thermal processes in single cells. As specific examples, the hybrid diamond nano-thermometer may
be applied to investigating the thermal conduction and dissipation of nanowires and 2D materials [39], to measuring fast (at millisecond timescale) temperature change during neuronal firing in neuron cells, and to detecting small thermal gradient (~μK across a cell) inside live cells in different live stages [4].

Methods

Experimental setup
A confocal-AFM correlation microscope was constructed to enable nano-manipulation of single copper-nickel alloy MNPs and in-situ temperature measurements (see Supplementary Figure 1). The AFM scanning head (BioScope Resolve, Bruker) was mounted on the confocal microscope to measure the topography and perform nanomanipulation of the MNP. The ODMR measurements were carried out using a home-built laser scanning confocal microscope. A 532 nm laser was adopted (MGL-III-532-200 mW, CNI) to excite the NV centers. An oil immersion objective lens (Nikon 100x 1.45NA) was used to collect the NV’s fluorescence signal, which was then detected by an avalanche photodiode (APD, SPCM-AQRH-15-FC, Excelitas) and counted by a data acquisition (DAQ, PCIe-6363, National Instruments). A microwave (MW) source (N5171B EXG Signal Generator, Keysight) and an amplifier (ZHL-16W-43-S+, Mini-Circuits) were used to generate microwave frequencies for spin measurements. A 20 μm copper wire was used to deliver MW. The sample temperature was controlled by a ceramic heater. The heating area of the heater is about 22 × 22 mm² and the pillar was placed in the center with diamond membrane size of 1 × 1 mm². Considering the excellent thermal conductivity of diamond material, we assumed the temperature was uniform across the diamond membrane (the distance between reference and hybrid sensor is also below 10 μm). For details see Supplementary Note.

NV centers in diamond pillars
In the experiments, high fluorescence intensity of single NV centers in diamond can enhance the sensitivity of the hybrid nanothermometer. Tapered nanopillar shape diamond waveguide was fabricated to achieve enhancement of the fluorescence collection efficiency of single NV centers in diamond. The fabrication process was developed and introduced by S. Momenzadeh et al. [30], where the diamond waveguide was fabricated by electron beam writing and reactive ion etching processes. The optical and spin coherent properties of the NV center are shown in Supplementary Figure 2. For details see Supplementary Note.

Temperature measurement and sensitivity estimation
The FID signal between the |m_τ = 0⟩ and |m_τ = −1⟩ state of the NV center is measured for the estimation of the temperature sensitivity. The method is illustrated in Fig. 2b. The FID signal, i.e., the photon count recorded at the end of the FID sequence is [34],

\[ S(t) \approx 1 - \frac{C}{2} + \frac{C}{2} \cos(2\pi ft) \exp \left[ -\left( \frac{t}{T_2^*} \right)^\nu \right], \]

where C is the contrast, \( f = f_r - f_p \) is the detuning of the transition frequency \( f_r \) from the frequency of the microwave pulses \( f_p \), \( T_2^* \) is the decoherence time and \( \nu \) is the exponent of the decay. Least-square fitting of the FID signal shown in Fig. 2b yields the parameters \( C = 0.27, \delta f = 2.7 \text{ MHz}, T_2^* = 1.8 \mu\text{s} \) and \( \nu = 3.3 \).
The optimal evolution time $t_e$ is determined by maximizing $|dS(t_e)/df_r|$. The FID signal variation depends on the temperature variation through

$$\delta S = \frac{dS(t_e)}{df_r} \times \frac{df_r}{dT} \times \delta T,$$

(3)

where $df_r/dT$ is the temperature susceptibility as shown in inset of Fig. 1d. Shot noise of the FID signal per unit measurement time is $\sigma \approx 1/\sqrt{L_{\text{eff}}}$, where $L_{\text{eff}}$ is the effective photon count rate (including only the photons recorded during the readout time, which occupies about 10% of the whole FID duty cycle in Fig. 2b). The shot-noise limited sensitivity of the hybrid nanothermometer is given by [34]

$$\eta_T = \sigma \left| \frac{dS(t_e)}{df_r} \right|^{-1} \left| \frac{df_r}{dT} \right|^{-1}.$$

(4)

Using the optimal temperature-susceptibility of the NV resonance frequency $df_r/dT = 47$ MHz K$^{-1}$ (see Fig. 1d) and the optimal evolution time $t_e = 1 \mu$s and using the effective count rate $L_{\text{eff}} = 9.6 \times 10^4$ s$^{-1}$ (corresponding to a saturated count rate $\sim 1 \times 10^6$ s$^{-1}$), we obtain the shot-limited temperature sensitivity to be $\eta_T \approx 76 \mu$K Hz$^{-1/2}$.

Associated content

Detailed description of the sample fabrication and experiment. This material is available free of charge in the supporting information.

Author information

R.B.L. and Q. L. conceived the idea and supervised the project. C.F.L., K. X., W.H.L., Q.L. and R.B.L. designed the experiments. K.X. and C.F.L. constructed the setup. C.F.L. and K.X. performed the experiments. W.H.L., C.F.L., K.X., Q.L. and R.B.L. analyzed the data. X.F. synthesized the magnetic nanoparticles. A.F., A. D. and J.W. implanted NV centers in diamond and fabricated the diamond nanopillars. C.F. L., W.H.L., K.X., Q.L. and R.B.L. wrote the paper and all authors commented on the paper.

The authors declare no competing financial interest.

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