Supplementary Materials for

Picotesla magnetometry of microwave fields with diamond sensors

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Section S1. Brief description of the detection scheme

S1.1 Derivation of rate equations

The Hamiltonian of the NV center with coupling to a microwave can be written as

\[ H = (D + \Delta_D)S_z^2 + (\Delta_B + \gamma_{NV}B_z)S_z + \gamma_{NV}b_1 \cos ftS_z, \]  

where \( S (S = 1) \) is the spin operator for the NV electron spin, \( D = 2.87 \text{ GHz} \) and \( \gamma_{NV} = -28.03 \text{ GHz/T} \) are the zero-field splitting and the gyromagnetic ratio of the NV electron spin, \( B_z \) is the axial component of the external static magnetic field, \( b_1 \) is the transverse magnetic field of the microwave with frequency \( f \), \( \Delta_D \) and \( \Delta_B \) are the frequency shifts induced by local strain or temperature fluctuations and magnetic field gradient, respectively. At resonance condition, for example, \( f = D + \gamma_{NV}B_z \), the Hamiltonian can be simplified to a time-independent form by moving to the interaction picture and omitting the high-frequency terms:

\[ H_{I} = \begin{pmatrix} \Delta & 0 & 0 \\ 0 & \frac{\Omega}{2} & 0 \\ 0 & 0 & 0 \end{pmatrix}, \]  

where \( \Omega = \gamma_{NV}b_1/\sqrt{2} \) is the corresponding Rabi frequency, and \( \Delta = \Delta_D + \Delta_B \). Then the evolution of NV center can be described by the master equation

\[ \dot{\rho} = -i[H_{I}, \rho] + \sum_{j=1}^{5} L_j \rho L_j^\dagger - \frac{1}{2}(L_j^\dagger L_j \rho + \rho L_j^\dagger L_j), \]  

where \( L_{1,2} = \sqrt{\Gamma_1/6}S_\pm \) and \( L_3 = \sqrt{2\Gamma_2}S_z \) are the longitudinal and transverse relaxation operators, respectively.

\[
L_4 = \sqrt{\Gamma_p} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix}, \quad L_5 = \sqrt{\Gamma_p} \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 0 & 0 \end{pmatrix}
\]

are the polarization operators. The master equation can be expanded as

\[
\begin{align*}
\rho'_{11} &= \frac{i\Omega}{2}(\rho_{12} - \rho_{21}) - \frac{\Gamma_1}{3}(\rho_{11} - \rho_{22}) - \Gamma_p \rho_{11}, \\
\rho'_{22} &= \frac{i\Omega}{2}(\rho_{21} - \rho_{12}) + \frac{\Gamma_1}{3}(\rho_{11} + \rho_{33} - 2\rho_{22}) + \Gamma_p (\rho_{11} + \rho_{33}), \\
\rho'_{33} &= -\frac{\Gamma_1}{3}(\rho_{33} - \rho_{22}) - \Gamma_p \rho_{33}, \\
\rho'_{12} &= -i\Delta \rho_{12} + \frac{i\Omega}{2}(\rho_{11} - \rho_{22}) - \Gamma_2 \rho_{12} + \frac{\Gamma_1}{3} \rho_{23}, \\
\rho'_{23} &= -\frac{i\Omega}{2} \rho_{13} - \Gamma_2 \rho_{23} + \frac{\Gamma_1}{3} \rho_{12}, \\
\rho'_{13} &= -i\Delta \rho_{13} - \frac{i\Omega}{2} \rho_{23} - 4\Gamma_2 \rho_{13}.
\end{align*}
\]  

(S4)
where the matrix components also satisfy $\rho_{11} + \rho_{22} + \rho_{33} \equiv 1$ and $\rho_{1i} = \rho_{i1}^\ast$. In the situation of $\Gamma_2 \gg \Gamma_1, \Gamma_p, \Omega$, the steady-state (i.e., $\rho = 0$) solution is

$$
\rho_1^\infty \approx \frac{(\Gamma_1 + 3\Gamma_p)(2\Gamma_1 + 3\Gamma_b)}{6(\Gamma_1 + \Gamma_p)(\Gamma_1 + 3\Gamma_p) + 9(\Gamma_1 + 2\Gamma_p)\Gamma_b},
$$
$$
\rho_{22}^\infty \approx \frac{\Gamma_1(2\Gamma_1 + 6\Gamma_p + 3\Gamma_b)}{6(\Gamma_1 + \Gamma_p)(\Gamma_1 + 3\Gamma_p) + 9(\Gamma_1 + 2\Gamma_p)\Gamma_b},
$$
$$
\rho_{33}^\infty \approx \frac{\Gamma_1(2\Gamma_1 + 6\Gamma_p + 3\Gamma_b)}{6(\Gamma_1 + \Gamma_p)(\Gamma_1 + 3\Gamma_p) + 9(\Gamma_1 + 2\Gamma_p)\Gamma_b},
$$

$$(S5)$$

Considering $\rho_{13}$ and $\rho_{23}$ also have a fast decay rate of $\Gamma_2$, $\rho_{13}$ and $\rho_{23}$ are negligible throughout the evolution, i.e.,

$$
\rho_{13} \approx 0,
$$
$$
\rho_{23} \approx 0.
$$

$$(S6)$$

The populations of NV states vary slowly comparing with $\Gamma_2$, which means $\rho_{11}$, $\rho_{22}$ and $\rho_{33}$ are quasi-static in the time scale of $\sim 1/\Gamma_2$, then we have

$$
\rho_{12}(t + \Delta t) \approx \frac{i\Omega}{2(\Gamma_2 + i\Delta)}(\rho_{11}(t) - \rho_{22}(t)) + \Delta \rho_{12} \cdot e^{-(\Gamma_2 + \Delta)t} \rightarrow \frac{i\Omega}{2(\Gamma_2 + i\Delta)}(\rho_{11}(t) - \rho_{22}(t)).
$$

$$(S7)$$

Substituting Eq. S6 and Eq. S7 into Eq. S4 yields

$$
P_1' = -\left(\frac{\Gamma_1}{3} + \frac{\Gamma_b}{2}\right)(P_1 - P_0) - \Gamma_p P_1,
$$
$$
P_0' = -\left(\frac{\Gamma_1}{3} + \frac{\Gamma_b}{2}\right)(P_0 - P_1) - \frac{\Gamma_1}{3}(P_0 - P_{-1}) + \Gamma_p(P_1 + P_{-1}),
$$
$$
P_{-1}' = -\left(\frac{\Gamma_1}{3}(P_{-1} - P_0) - \Gamma_p P_{-1}.
$$

$$(S8)$$

It consists with the simple relaxation picture as shown in Fig. S1a. Similarly, one can directly written the rate equations of a two-level system (Fig. S1b) as discussed in the main text.

![FIG. S1: Simple relaxation picture of three- and two-level systems.](image-url)
As described in the main text, the analysis of two-microwave case is similar with single-microwave case by substituting $\Gamma_b \rightarrow \Gamma_{\text{aux}} + 2\sqrt{\Gamma_{\text{aux}}\Gamma_b}\cos(\delta t + \phi)$ into the above rate equations Eq. S8. After some simplifications, the rate equations becomes

$$
P'_0 = -\left[\Gamma_1 + \Gamma_p + \frac{\Gamma_{\text{aux}}}{2} + \sqrt{\Gamma_{\text{aux}}\Gamma_b}\cos(\delta t + \phi)\right] P_0 + \left[\frac{\Gamma_{\text{aux}}}{2} + \sqrt{\Gamma_{\text{aux}}\Gamma_b}\cos(\delta t + \phi)\right] P_1 + \frac{\Gamma_1}{3} + \Gamma_p,
$$

$$
P'_1 = -\left[\frac{\Gamma_1}{3} + \Gamma_p + \frac{\Gamma_{\text{aux}}}{2} + \sqrt{\Gamma_{\text{aux}}\Gamma_b}\cos(\delta t + \phi)\right] P_1 + \left[\frac{\Gamma_1}{3} + \frac{\Gamma_{\text{aux}}}{2} + \sqrt{\Gamma_{\text{aux}}\Gamma_b}\cos(\delta t + \phi)\right] P_0.
$$

(S9)

If only considering the situation of $\Gamma_b \ll \Gamma_{\text{aux}}$, the quasi-steady-state solution should be a perturbation of $P^\infty$, i.e., $P^\infty(t) \approx P^\infty + A_0\cos(\delta t + \varphi_0) + \cdots$ Substituting the trail solution

$$
P^\infty_0(t) \approx P^\infty_0 + A_0\cos(\delta t + \varphi_0),
$$

$$
P^\infty_1(t) \approx P^\infty_1 + A_1\cos(\delta t + \varphi_1),
$$

into Eq. S9 and only preserving terms with frequency $\delta$ yields

$$-A_0\delta \sin(\delta t + \varphi_0) = -(\Gamma_1 + \Gamma_p + \frac{\Gamma_{\text{aux}}}{2})A_0\cos(\delta t + \varphi_0) + \frac{\Gamma_{\text{aux}}}{2}A_1\cos(\delta t + \varphi_1) + \sqrt{\Gamma_{\text{aux}}\Gamma_b}(P^\infty_1 - P^\infty_0)\cos(\delta t + \phi),
$$

$$-A_1\delta \sin(\delta t + \varphi_1) = (\frac{\Gamma_1}{3} + \frac{\Gamma_{\text{aux}}}{2})A_0\cos(\delta t + \varphi_0) - (\frac{\Gamma_1}{3} + \Gamma_p + \frac{\Gamma_{\text{aux}}}{2})A_1\cos(\delta t + \varphi_1) + \sqrt{\Gamma_{\text{aux}}\Gamma_b}(P^\infty_0 - P^\infty_1)\cos(\delta t + \phi).
$$

(S10)

Adding the two equations yields

$$A_0\sqrt{\frac{2}{3}\Gamma_1 + \Gamma_p}^2 + \delta^2\cos(\delta t + \varphi_0 + \theta_0) = -A_1\sqrt{\frac{1}{3}(\Gamma_1 + \Gamma_p)^2 + \delta^2}\cos(\delta t + \varphi_1 + \theta_1),
$$

(S11)

where $\theta_0 = \arctan\frac{\delta}{\frac{2}{3}\Gamma_1 + \Gamma_p}$ and $\theta_1 = \arctan\frac{\delta}{\frac{2}{3}\Gamma_1 + \Gamma_p}$. It is equivalent to

$$A_1 = \sqrt{\frac{\frac{2}{3}(\Gamma_1 + \Gamma_p)^2 + \delta^2}{\frac{1}{3}(\Gamma_1 + \Gamma_p)^2 + \delta^2}}A_0 = \eta \cdot A_0,
$$

$$\varphi_1 = \pi + \varphi_0 + \theta_0 - \theta_1 = \pi + \varphi_0 + \Delta \theta.
$$

(S12)

Substituting Eq. S12 into Eq. S10 yields

$$A_0 = \frac{(P^\infty_0 - P^\infty_1)\sqrt{\Gamma_{\text{aux}}\Gamma_b}}{\sqrt{(\Gamma_1 + \Gamma_p + \frac{1}{2}\Gamma_{\text{aux}})^2 + \delta^2 + \frac{1}{4}\eta^2\Gamma_{\text{aux}}^2 + \eta\Gamma_{\text{aux}}[(\Gamma_1 + \Gamma_p + \frac{1}{2}\Gamma_{\text{aux}})\cos\Delta \theta - \delta \sin\Delta \theta]}},
$$

(S13)

where

$$P^\infty_0 - P^\infty_1 = \frac{(\frac{1}{2}\Gamma_1 + \Gamma_p)\Gamma_p}{(\Gamma_1 + \Gamma_p)(\frac{1}{2}\Gamma_1 + \Gamma_p) + (\frac{1}{2}\Gamma_1 + \Gamma_p)\Gamma_{\text{aux}}}.
$$

Although this formula looks complicated, in the situation of $\Gamma_1 \ll \Gamma_p$, it can be simplified as

$$A_0 \approx \frac{\Gamma_p\sqrt{\Gamma_{\text{aux}}\Gamma_b}}{(\Gamma_p + \Gamma_1 + \Gamma_{\text{aux}})(\Gamma_p + \Gamma_1 + \Gamma_{\text{aux}})^2 + \delta^2},
$$

(S14)

which is the same as the two-level system discussed in the main text. In our experiment, the difference between these two formulas is smaller than 5%.
S2. Measurements of basic experimental parameters

S2.1 Transitional relaxation rate

As discussed in the main text and shown in Fig. S2A, the transitional relaxation is induced by two kinds of noise: static or quasi-static noise and fast fluctuating noise. The former comes from the inhomogeneity of local strain and magnetic fields or temperature fluctuations, resulting an energy shift of $\Delta$. The latter comes from the surrounding bath spins, resulting a decoherence process with rate of $\Gamma_2$. To eliminate the static and quasi-static noises, we perform a measurement of Hahn echo. The residual noise is captured by an Ornstein-Uhlenbeck process with variance of $b^2$ and correlation time of $\tau_c$. In this simple model, the Hahn echo signal has analytic solution

$$S_{\text{Hahn, single}}(t) = \frac{1}{2} + \frac{1}{2} \exp \left[ -b^2 \tau_c^2 \left( \frac{t}{\tau_c} - 3 - e^{-t/\tau_c} + 4e^{-t/\tau_c} \right) \right].$$

(S15)

Different NV centers experience different noise, of which the probability density function (PDF) is

$$P(b) = \frac{2}{\pi} \frac{1}{b^2} e^{-\frac{\sigma^2}{2b^2}}.$$

(S16)

After ensemble average, it becomes

$$S_{\text{Hahn, ensemble}}(t) = \int_{-\infty}^{+\infty} S_{\text{Hahn, single}}(t) P(b) db$$

$$= \frac{1}{2} + \frac{1}{2} \exp \left[ -2\sigma^2 \tau_c^2 \left( \frac{t}{\tau_c} - 3 - e^{-t/\tau_c} + 4e^{-t/\tau_c} \right) \right].$$

(S17)

Substituting $\sigma^2 = \sigma^2 \tau_c/2$ into Eq. S17 yields

$$S_{\text{Hahn, ensemble}}(t) = \frac{1}{2} + \frac{1}{2} \exp \left[ -4\sigma^2 \tau_c \left( \frac{t}{\tau_c} - 3 - e^{-t/\tau_c} + 4e^{-t/\tau_c} \right) \right].$$

(S18)

This formula can fit the experimental results well with $\sigma_2 = 8 \pm 4$ kHz and $\tau_c = 4 \pm 2$ $\mu$s (Fig. S2B). Now we get the PDF of $\Gamma_2 (= b^2 \tau_c)$

$$P(\Gamma_2) = \frac{\sqrt{2\pi}\gamma_0^2}{\Gamma_2^2} b^2 e^{-\frac{\sigma_2^2}{2\Gamma_2^2}}.$$ 

(S19)

We then consider the (quasi-)static noise, which dominates the line broadening of ODMR spectrum. As given by Eq. 5 in the main text, the ODMR spectrum can be calculated as

$$\langle \Delta P_0^{\infty}(\nu) \rangle = \int \int \frac{\gamma_0^2 b^2 \Gamma_0 \Gamma_2}{4(\Gamma_0 + \Gamma_1)^2[\Gamma_2^2 + (\nu + \Delta)^2]} P(\Delta) P(\Gamma_2) d\Delta d\Gamma_2$$

(S20)

where $\Delta$ obeys normal distribution with PDF of

$$P(\Delta) = \frac{1}{\sqrt{2\pi}\sigma_1} e^{-\frac{\Delta^2}{2\sigma_1^2}}.$$ 

(S21)

The experimental result can be well fitted with $\sigma_1 \sim 200$ kHz (Fig. S2C). The measured responsivity can be well explained by this model and the two parameters $\sigma_{1,2}$. If phenomenologically attributing the transitional relaxation only to the dephasing process, i.e., $\Gamma_b = \gamma_0^2 b^2 / 2\Gamma_2^2$, where $\Gamma_2^2 \sim 240$ kHz can be estimated from the linewidth of the ODMR spectrum, the calculated responsivity will show obvious deviation from the measurement (Fig. S2D). Nevertheless, it can roughly estimate the maximal responsivity up to a mere constant.
FIG. S2: Measurements of transitional relaxation. A Schematic of the microscopic mechanism of noise. B Hahn echo measurement of NV ensemble. C Zoom in on the central peak of the ODMR spectrum. D Comparison of the two-noise model and the phenomenological model.

### S2.2 Intrinsic longitudinal relaxation rate and polarization rate

The solution of the rate equations Eq. S8 is

\[ P_0(t) = P_0^\infty + C_1 e^{-K_1 t} + C_2 e^{-K_2 t}, \]  

(S22)

where

\[ K_{1,2} = \frac{2\Gamma_1}{3} + \Gamma_p + \frac{\Gamma_b}{2} \pm \sqrt{\frac{\Gamma_1^2}{9} + \frac{\Gamma_p^2}{4} + \frac{\Gamma_1 \Gamma_b}{6}}, \]

and \( C_{1,2} \) are constants depending on the initial state. When the microwave is turned off (\( \Gamma_b = 0 \)), the solution has a single-exponential decay form

\[ P_0(t) = \frac{\Gamma_1 + 3\Gamma_p}{3\Gamma_1 + 3\Gamma_p} e^{-(\Gamma_1 + \Gamma_p)t}. \]  

(S23)

Therefore, \( \Gamma_1 \) and \( \Gamma_p \) can be extracted from the revival rate of the fluorescence (Fig. S3).
FIG. S3: Measurements of longitudinal relaxation. A The fluorescence will decay when the microwave is turned on, and revival when the microwave is turned off. B The revival rate shows linear dependence on $P_L$. The intercept of the linear fit gives $\Gamma_1 = 102 \pm 5$ Hz, while the slope is $250 \pm 6$ Hz/W.

S2.3 Benchmark of microwave field strength

The microwave is radiated from a loop antenna, and the field strength $b_1 = k\sqrt{P_{MW}}$, where $P_{MW}$ is the input microwave power. When it is strong enough ($P_{MW} = 6.1$ W), the time trace of fluorescence shows Rabi oscillations (Fig. S4A). The Rabi frequency $\Omega = \gamma_N V b_1 / \sqrt{2}$ shows a linear relationship to $\sqrt{P_{MW}}$ with a proportionality coefficient of $225 \pm 3$ kHz/$\sqrt{W}$. Therefore, the proportionality coefficient $k = 11.3 \pm 0.2$ µT/$\sqrt{W}$. Here $P_{MW}$ is calibrated by a microwave analyzer (Keysight N9917A).

FIG. S4: Rabi oscillation. A Rabi oscillation under different input microwave power. Points are experimental results, while lines are sine damping fit. B Dependence of Rabi frequency on microwave power. The fitted slope is $225 \pm 3$ kHz/$\sqrt{W}$. 
Section S3. Measurements with different laser power

S3.1 Optimization of laser power

According to Eq. S14, the oscillation amplitude $A_0$ has similar dependence on $\Gamma_{aux}$ and $\Gamma_p$. Assuming both of them are proportional to $\Gamma_x$, and considering the fluorescence is proportional to $\Gamma_p$, the signal is

$$S \propto \Gamma_x^{2.5} (\Gamma_x + \alpha \Gamma_1)^2.$$

(S24)

If the noise is dominated by photon shot noise ($\propto \Gamma_p^{0.5}$), then shot-noise-limited signal to noise ratio (SNR) is

$$\text{SNR} \propto \frac{\Gamma_x^{2}}{(\Gamma_x + \alpha \Gamma_1)^2}.$$

(S25)

which is positively correlated with $\Gamma_x$ ($\Gamma_{aux}$, $\Gamma_p$) (Fig. S5A). However, the main noise in our experiment is laser-induced photon noise, which is $\propto \Gamma_p$ (Fig. S5B). So that

$$\text{SNR} \propto \frac{\Gamma_x^{1.5}}{(\Gamma_x + \alpha \Gamma_1)^2},$$

(S26)

indicating the existence of an optimal $\Gamma_x$. To find the optimal point, we repeat the measurements under different microwave and laser power (Fig. S5C). As shown in Fig. S5D, the dependence of SNR on $P_L$ can be extract from Fig. S5B, C. Here we choose 0.8 W instead of 1 W as the optimal laser power because of the laser heating problem, as we will discuss latter.

FIG. S5: Optimal laser power. A Shot-noise-limited SNR. B Dependence of noise on laser power. Points are experimental results, which are measured by the RMS of the Fourier transform spectrum around 480 Hz with a span of 0.1 Hz. Two dot lines are indications of linear and square-root scaling. C Dependence of signal on laser and microwave power. Dash line indicates the points to extract SNR. D Experimental SNR under different laser power.
High-power laser will heat the diamond sensor, and thus induces various problems, such as increases of relaxation rate, drifts of resonant frequencies, and mechanical instabilities of the setup. We monitor the resonant frequencies under the continuous illumination with different laser power (Fig. S6A). Since the frequency drift $\Delta f$ is proportional to the temperature variation $\Delta T$, and $\Delta f/\Delta T \approx -77$ kHz/K, the temperature increase induced by laser heating can be extracted from the ODMR spectrum, as shown in Fig. S6B.

FIG. S6: Laser heating. A ODMR spectrum of NV centers on different laser power. B The heating shows a linear dependence on laser power $P_L$, where the fitted slope is $k = 39.3 \pm 0.7$ K/W.
The total noise consists of the laser-induced noise $n_{\text{laser}}$, the photon shot noise $n_{\text{shot}}$, and the electric noise of the detection system $n_{\text{electric}}$. Among them, the photon shot noise can be independently calculated from the fluorescence. In our experiment, the fluorescence power is 1.6 mW under the laser power of $P_L = 0.8$ W. The photon shot noise is

$$n_{\text{shot}} = 2\sqrt{2eI_{\text{fl}} \cdot G} \approx 24 \mu V/\sqrt{\text{Hz}}. \quad (S27)$$

where $e = 1.602 \times 10^{-19}$ C is the electron charge, $I_{\text{fl}} \approx P_L \cdot 0.5$ A/W is the photon current, $G = 750$ kV/A is the total transimpedance gain, the coefficient 2 comes from the definition of noise (RMS of the base line in Fourier transform spectra) in our measurement. For arbitrary laser power $P_L$, the photon shot noise is

$$n_{\text{shot}} = \sqrt{\xi \cdot P_L}. \quad (S28)$$

where $\xi = 720 \mu V^2 \cdot W^{-1} \cdot \text{Hz}^{-1}$. Considering the laser-induced noise is proportional to $P_L$ and the electric noise is independent of $P_L$, the total noise can be written as

$$n_{\text{total}} = \sqrt{n_{\text{laser}}^2 + n_{\text{shot}}^2 + n_{\text{electric}}^2} = \sqrt{\chi \cdot P_L^2 + \xi \cdot P_L + n_{\text{electric}}^2}. \quad (S29)$$

In order to extract the contribution of each noise, we measure the total noise under different $P_L$, and used Eq. S29 to fit the $n_{\text{total}}$-$P_L$ curve. Figure S7 give the extracted noises at $P_L = 0.8$ W, where the total noise is indeed dominated by the laser-induced noise in most of the frequency range. Specifically, the laser-induced noise and electric noise are $33 \mu V/\sqrt{\text{Hz}}$ and $14 \mu V/\sqrt{\text{Hz}}$, respectively, at the optimal point ($P_L = 0.8$ W, $\delta = 480$ Hz).

---

**FIG. S7**: **Noise contribution with laser power of 0.8 W.**

A: Extracted noise contribution. The photon-shot noise is white with known calculated value. The laser-induced noise and electric noise are fitted by Eq. S29. Here the total noise is experimental result, so it is smaller than the laser-induced noise at some frequency band due to imperfect fits.

B: The corresponding goodness of fit.
As shown in Fig. S8, we repeat the measurement of responsivity on different days. The dependence on $B_1$ remains the same up to an overall change in intensity. So we attribute this change to the variation of fluorescence contrast, which is induced by slow dynamics of the NV charge states.

**FIG. S8:** Repeated measurements of responsivity on different days.