Supplementary material to "Cavity-enhanced room-temperature magnetometry using absorption by nitrogen-vacancy centers in diamond"

K. Jensen,[1] N. Leefer,[1] A. Jarmola,[1] Y. Dumeige,[2,3] V. M. Acosta,[1] P. Kehayias,[1] B. Patton,[1,4] and D. Budker[1]

[1]*Department of Physics, University of California, Berkeley, California 94720-7300, USA*
[2]*UEB, Université Européenne de Bretagne, Université de Rennes I*
[3]*CNRS, UMR 6082 FOTON, Enssat, 6 rue de Kerampont, CS 80518, 22305 Lannion cedex, France*
[4]*Physik-Department, Technische Universität München, 85748 Garching, Germany*

CAVITY TRANSMISSION

Consider an optical cavity consisting of two identical mirrors with intensity transmission and reflection coefficients T and R such that $T+R=1$. An absorptive element with a single-pass intensity transmission coefficent L is placed inside the cavity. The intensity transmission through the cavity equals [S1]

$$I/I_0 = \frac{T^2 L}{(1-RL)^2 + 4RL\,(\sin\phi)^2}, \tag{S1}$$

where I_0 is the input intensity and ϕ is the single-pass phase aquired by the light inside the cavity. In our experiment the mirrors are curved, and the input light is matched to the TEM_{00} cavity mode. When scannning the frequency of the light, maxima in the transmission will be spaced by the frequency interval $c/(2l_{\mathrm{opt}})$, where l_{opt} is the optical path length of the cavity and c the speed of light. The maximum transmission equals

$$I_{\max}/I_0 = \frac{T^2 L}{(1-RL)^2}. \tag{S2}$$

The finesse $\mathcal{F}$ of the cavity is defined as the frequency spacing $c/(2l_{\mathrm{opt}})$ divided by the cavity resonance FWHM $\Delta\nu_c$ and is given by:

$$\mathcal{F} = [c/(2l_{\mathrm{opt}})]/\Delta\nu_c \approx \pi\frac{\sqrt{RL}}{1-RL}. \tag{S3}$$

We are interested in measuring the loss inside the cavity. Suppose there is a small change $|\delta L| = L_0 - L$ in the parameter L from its initial value L_0. By Taylor expanding Eq. (S2) we find the change in the cavity transmission $\Delta I = I_{\max}(L=L_0) - I_{\max}(L = L0 - |\delta L|)$ to be

$$\frac{\Delta I}{I_{\max}(L=L_0)} = \frac{(1+RL_0)}{(1-RL_0)}\frac{|\delta L|}{L_0} \approx \frac{2\mathcal{F}}{\pi}\frac{|\delta L|}{L_0}. \tag{S4}$$

We see that the sensitivity to a small change in absorption is increased by the factor $2\mathcal{F}/\pi$ by employing a cavity compared to a single pass where $\Delta I/I(L=L_0) \approx |\delta L|/L_0$. The effective number of passes through the absorptive element inside the cavity is therefore $2\mathcal{F}/\pi$.

BIREFRINGENCE

Assume the cavity contains a diamond with thickness d and index of refraction $n = 2.4$ such that the total length of the cavity is $l+d$ and the optical path length of the cavity is $l_{\mathrm{opt}} = l + nd$. Light input to the cavity will be resonantly transmitted if the frequency of the light equals a cavity resonance frequency:

$$\nu_m = m\cdot\frac{c}{2(l+nd)} + \nu_G, \tag{S5}$$

where $m = 0, 1, 2, ...$ and ν_G is related to the Gouy phase shift of a Gaussian beam [S1]. We now assume that the diamond is birefringent such that two orthogonal linear polarizations, here denoted H and V, have slightly different index of refraction: $n_H = n$ and $n_V = n + \Delta n$. The resonance frequencies for H and V polarizations differ by the amount

$$\nu_m^H - \nu_k^V = \frac{m-k}{k}\cdot\nu_k^V + \frac{\Delta n\cdot d}{l+(n+\Delta n)\,d}\cdot\nu_m^H. \tag{S6}$$

The frequency difference $\nu_m^H - \nu_k^V$ for H and V polarized light is measured in the experiment [see trace 3 in Fig. 2(a)]. We believe that neighboring resonances for H and V polarized light have the same fringe order $m = k$. This is based on the observation that by moving the diamond (and thereby probing different locations on the diamond which have different degrees of birefringence), neighboring H and V resonances can be made to overlap but not cross. Assuming $m = k$ and $\Delta n \ll n$ we find (dropping the subscripts)

$$\Delta\nu \equiv \nu^H - \nu^V \approx \frac{\Delta n\cdot d}{l+nd}\cdot\nu_0, \tag{S7}$$

where ν_0 is the laser frequency. This equation can be used to estimate the degree of birefringence Δn of the diamond.

We observe significant birefringence in all our diamond samples and find that the degree of birefringence depends on the spot on the diamond. The birefringence is either an intrinsic property of the diamond [S2] or due to stress induced by the mount. Traces 2 and 3 in Fig. 2(a) show the cavity transmission for two different polarizations of

the light input to the cavity. When the input polarization is aligned either along or perpendicular to the diamond's axis of birefringence (trace 2), peaks separated by the $c/(2l_{\rm opt}) = 3.0$ GHz are observed. If the polarization is neither parallel nor perpendicular to the birefringence axis (trace 3), twice the number of resonances is observed since light polarized parallel and perpendicular to the material's birefringence axis experiences different index of refraction. The degree of birefringence, Δn, can be estimated from the frequency difference of two neighbouring resonances $\Delta\nu$ using Eq. (S7). From trace 3 we find $\Delta\nu = 70$ MHz, which leads to a difference in index of refraction $|\Delta n| = 6.1\cdot 10^{-5}$, where we used $\nu_0 = c/\lambda = 2.88\cdot 10^{14}$ Hz and $d = 0.2$ mm. This degree of birefringence corresponds to a phase-shift difference between the two polarizations of $|\Delta\phi| = (2\pi/\lambda)\,|\Delta n|\,d = 4.2$ deg.

ABSORPTION BY NV CENTERS

NV centers are excited from the spin-triplet ground state $^3A_2$ to the spin-triplet excited state ^3E by 532 nm pump light. For small pump light intensities I_P where the transition is not saturated, the excitation rate Γ_P is

$$\Gamma_P = \sigma_P \cdot \Phi_P = \sigma_P \cdot \frac{I_P}{hc/\lambda_P}, \tag{S8}$$

where σ_P is the cross-section for the pump light on the $^3A_2 \leftrightarrow {}^3E$ transition, Φ_P is the flux of pump photons, h is Planck's constant, and λ_P is the pump wavelength. The condition for saturating the $^3A_2 \leftrightarrow {}^3E$ transition is $\Gamma_P^{\rm sat,triplet} \approx A_{21}$, where A_{21} is the inverse lifetime of the ^3E excited state. From this condition we calculate the saturation intensity to be

$$I_P^{\rm sat,triplet} \approx \frac{A_{21}}{\sigma_P}\cdot\frac{hc}{\lambda_P} \sim 10\ {\rm GW/m^2}, \tag{S9}$$

where we have used the values $A_{21} = 1/(12\ {\rm ns})$ [S3], $\sigma_P = 3\cdot 10^{-21}$ m^2 [S4], and $\lambda_P = 532$ nm.

An NV center can decay from the spin-triplet excited state ^3E to the upper spin-singlet state $^1A_1$ from which it decays in < 1 ns [S5] to the lower metastable spin-singlet state ^1E. The NV center will therefore mainly populate the lower spin-singlet state under strong optical pumping. The saturation pump power for populating the singlet state can be calculated using a rate-equation model for the spin-state dynamics of the NV center [S6]. An order of magnitude estimate can be obtained from the condition $\Gamma_P^{\rm sat,singlet} \approx \Gamma_s$, where $\Gamma_s = 1/(200\ {\rm ns})$ is the inverse lifetime of the metastable singlet state [S5]. The saturation intensity is then

$$I_P^{\rm sat,singlet} \approx \frac{\Gamma_s}{\sigma_P}\cdot\frac{hc}{\lambda_P} = \frac{\Gamma_s}{A_{21}}\cdot I_P^{\rm sat,triplet} = 600\ {\rm MW/m^2}. \tag{S10}$$

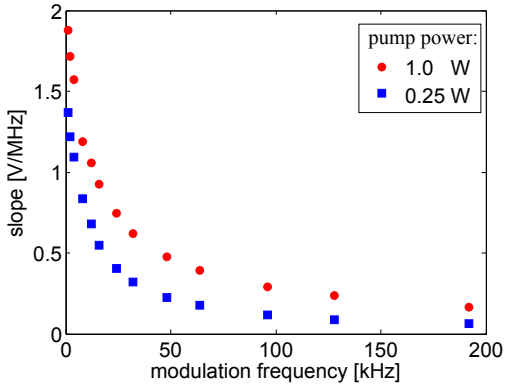

FIG. S1: The slope α as a function of the modulation frequency for two pump-light powers (1.0 W and 0.25 W measured before the cavity).

The population in the metastable singlet state as a function of pump intensity can be estimated as

$$p_s \approx \frac{I_P}{I_P + I_P^{\rm sat,singlet}}. \tag{S11}$$

We now consider a diamond with a density of NV centers $n_{\rm NV}$ and thickness d. If we assume that all NV centers experience the same pump intensity I_P, the amount of absorption is

$$A = \sigma_{\rm IR}\cdot n_{\rm NV}\cdot d\cdot p_s = \sigma_{\rm IR}\cdot n_{\rm NV}\cdot d\cdot\frac{I_P}{I_P + I_P^{\rm sat,singlet}}, \tag{S12}$$

where $\sigma_{\rm IR}$ is the cross-section for the $^1E \leftrightarrow {}^1A_1$ transition. In our experiment the pump intensity is not homogeneous inside the diamond since the pump light has a Gaussian transverse intensity profile and because the pump light is attenuated while passing through the diamond due to absorption by NV centers. However, for simplicity, we assume a homogeneous pump intensity in our model used for fitting the experimental data shown Fig. 2(b) for the cavity transmission as a function of pump power.

RESPONSE OF THE NV CENTERS

Using the modulation technique described in the main text, we obtain a magnetometer signal $S_{\rm LI} \approx \alpha\,(f_c - f_{\rm res})$ which is linear in $f_c - f_{\rm res}$ close to resonance. Figure S1 shows the slope α for different modulation frequencies. The slope decreases with increasing modulation frequency because of the finite response time of NV centers to changes in the magnetic field [S7]. For instance, the slope has decreased by a factor of two for modulation frequencies 13.5 kHz and 10.6 kHz for the pump powers 1.0 W and 0.25 W, respectively. Those powers correspond to peak intensitites 80 MW/m^2 and 20 MW/m^2,

respectively (with a 90 μm beam waist). The response time limits the magnetometer bandwidth. Faster response times and higher bandwidths can be achieved by increasing the pump intensity, for instance, in Ref. [S7], a bandwidth of a few MHz was achieved with a pump intensity of 20 GW/m^2. The maximum bandwidth of IR-absorption based magnetometry in diamond is limited by the metastable singlet state lifetime to a few MHz [S8].

[S1] Peter W. Milonni and Joseph H. Eberly. *Lasers*. Wiley, 1988.

[S2] I. Friel, S.L. Clewes, H.K. Dhillon, N. Perkins, D.J. Twitchen, and G.A. Scarsbrook. *Diamond and Related Materials*, 18:808, 2009.

[S3] A. Batalov, C. Zierl, T. Gaebel, P. Neumann, I.-Y. Chan, G. Balasubramanian, P. R. Hemmer, F. Jelezko, and J. Wrachtrup. *Phys. Rev. Lett.*, 100:077401, 2008.

[S4] Tse-Luen Wee, Yan-Kai Tzeng, Chau-Chung Han, Huan-Cheng Chang, Wunshain Fann, Jui-Hung Hsu, Kuan-Ming Chen, and Yueh-Chung Yu. *The Journal of Physical Chemistry A*, 111:9379, 2007.

[S5] V. M. Acosta, A. Jarmola, E. Bauch, and D. Budker. *Phys. Rev. B*, 82:201202, 2010.

[S6] L. Robledo, H. Bernien, T. van der Sar, and R. Hanson. *New Journal of Physics*, 13:025013, 2011.

[S7] C. S. Shin, C. E. Avalos, M. C. Butler, D. R. Trease, S. J. Seltzer, J. P. Mustonen, D. J. Kennedy, V. M. Acosta, D. Budker, A. Pines, and V. S. Bajaj. *Journal of Applied Physics*, 112:124519, 2012.

[S8] V. M. Acosta, E. Bauch, A. Jarmola, L. J. Zipp, M. P. Ledbetter, and D. Budker. *Appl. Phys. Lett.*, 97:174104, 2010.