Room Temperature Operation of a Radiofrequency Diamond Magnetometer near the Shot Noise Limit

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Abstract: We operate a nitrogen vacancy (NV) diamond magnetometer at ambient temperatures and study the dependence of its bandwidth on experimental parameters including optical and microwave excitation powers. We introduce an analytical theory that yields an explicit formula for the response of an ensemble of NV spins to an oscillating magnetic field, such as in NMR applications. We measure a detection bandwidth of 1.6 MHz and a sensitivity of 4.6 nT/Hz, unprecedented in a detector with this active volume and close to the photon shot noise limit of our experiment.

The negatively charged nitrogen vacancy center (NV−), a substitutional point defect in diamond, exhibits favorable optical and magnetic properties that have recently been exploited in several applications. For example, their brightness, optical stability, and biological inertness make NV− defect-harborin nanodiamonds ideal probes in bioimaging [1] and fluorescence resonance energy transfer [2] experiments. More importantly, the NV− defect forms a magneto-optical system whose spin state can be initialized and read out optically. Because the NV spin-coherence lifetimes can be as long as milliseconds in an isotopically pure diamond lattice [3], the system is an ideal platform for experimental quantum information science. Among such devices are precision magnetometers that have applications as industrial sensors, probes of magnetic materials, and as detectors of magnetic resonance. Practical magnetic field sensors for electron spin resonance (ESR), nuclear magnetic resonance (NMR), and other similar applications must sensitively detect weak, oscillating magnetic fields whose frequency and bandwidth cannot be arbitrarily controlled. Thus, transient response of an ensemble of NV− centers, characterized by its sensitivity to magnetic fields oscillating over a wide bandwidth, is a critical metric for applications in NMR and magnetic resonance imaging.

To quantitatively understand the transient dynamics that limit the bandwidth in a diamond magnetometer, we have developed an analytical treatment of the transient response of an NV− ensemble under microwave and optical irradiation. The NV− center ground state is a spin triplet (S=1) with zero field splitting of 2.87 GHz in sublevels, i.e. m_s = 0 and m_s = ±1. A static magnetic field shifts the transition between m_s = 0 and m_s = ±1 out of resonance with the microwave field. The system can thus be modeled as a set of three two-level systems, whose resonant frequencies are separated by ~2.1 MHz due to the hyperfine coupling between the electron spin and the 14N nuclear spin. Since the three two-level systems can be considered essentially isolated from each other [4,SI], Bloch equations can be used to model the response of each system individually, with the optically-induced spin-relaxations included in the model only through their contributions to T_1 and T_2, where T_1 and T_2 are spin-lattice relaxation time, and spin-spin relaxation time, respectively. As shown in the Supporting Information, the response time, τ, of a two-level system to an oscillating field during continuous excitation is a weighted average of 1/T_1 and 1/T_2:

\[ 1/\tau = \frac{(\omega_0 \cos \phi)^2 / T_1 + \Delta^2 / T_2}{(\omega_0 \cos \phi)^2 + \Delta^2} \]  

where Δ is the offset from resonance of the microwave field, \( \omega_0 = B_0 / 2 \) is the Rabi frequency at resonance where 2π is the electron gyromagnetic ratio, \( B_1 \) is the amplitude of the
linearly polarized time-varying magnetic field, and the angle $\phi$ is the solution to the equation

$$
\tan \phi = \frac{\Delta(1/T_1 - 1/T_2)}{(\alpha_1 \cos \phi)^2 + \Delta^2}. \tag{2}
$$

(In the limit where the Rabi frequency $\sqrt{\alpha_1^2 + \Delta^2} \gg 1/T_1, 1/T_2$, we have $\cos \phi = 1$, while $\cos \phi = 0.5$ at low microwave power and high laser power.)

Our goal is to develop sensitive magnetometers that can be integrated with microfabricated, microfluidic NMR devices [5], in which the signal is a sum of damped oscillations with up to ~100 kHz bandwidth and ~1-3 nT amplitude, and the field from the sample dies off within 10-50 microns from the surface, limiting the detector’s volume. The experiments we present below validate our analytical theory for magnetometer sensitivity and bandwidth and demonstrate that these specifications can be achieved in a practical device. We first explore the frequency response of the detector, demonstrating a bandwidth of more than 1.6 MHz. For the sensitivity experiments, we use a continuous-wave, single modulation technique in which the oscillating magnetic field itself modulates the fluorescence signal and is detected by lock-in or Fourier methods; in combination with gradiometric detection to remove technical noise, this allows us to achieve a sensitivity of ~4.6 nT/Hz at room temperature, with an active spot size of only ~1 $\mu$m, matched to the size of small microfluidic channels and much smaller than is practical with inductive NMR detection techniques.

For comparison, others have reported sensitivity of ~4 nT/Hz using a single NV center [3] or ~20 nT/Hz using an ensemble of NV centers, both at ambient temperatures [6]. Infrared-absorption detection, using an NV ensemble, achieved a sensitivity of ~7 nT/Hz at 45 K [7]. In that work, a theoretical bandwidth of a few MHz was suggested, but experimental operation was limited to a few hundred Hz. With a single NV center, a detection bandwidth of a several hundred kHz has been reported [8].

The diamond sample in our experiments is S9 (NV$^-$ concentration of ~2 ppm) described in Ref. [9], and the geometry of our experiment is illustrated in Fig. 1, and described further in the caption. For optically-detected ESR experiments, a static magnetic field of ~20 G was applied to break the orientational degeneracy of the NV center, and the experiments generally probed a single manifold of resolved hyperfine lines. We estimated the $T_2^*$ from the first derivative of the absorption spectra to be 130±5 ns [SI]. The frequency response of the NV ensemble at a given optical and microwave power was obtained by frequency-modulating the microwaves at a modulation rate $f_{\text{mod}}$ centered at $f_{\text{mic}}$. The slope of the absorption spectrum was maximum about this frequency, as schematically shown in the inset of Fig. 1. Representative data together with simulations are shown in Fig. 2.
ensemble $T_1$ is $\sim 152.0 \mu s$ and $T_2$ is $\sim 1.5 \mu s$, decreasing to 2.2 \mu s and 140 ns, respectively, at 40 mW of optical excitation power. While this qualitatively explains the increase in bandwidth with optical power, we note that the actual dynamics of the ensemble is complicated by the Gaussian profile of the excitation spot, which introduces a significant spatial inhomogeneity in $T_1$ and $T_2$.

Importantly, these data are explained by our analytical theory in the regime where it applies to the experiment. Bloch-equation simulations that used measured parameters are in quantitative agreement with experimental bandwidth data at low microwave powers ($\omega_\delta/2\pi \sim 0.1$ MHz). For laser powers near 1 mW, the simulated curves also showed qualitative agreement with the bandwidth data over the full range of microwave powers, but for laser power $\geq 6$ mW and $\omega_\delta/2\pi \geq 1$ MHz, simulations that used measured parameters were qualitatively different than experimental curves. The measured time constants, $T_1$ and $T_2$, changed dramatically when the laser power increased from 1 mW to 6 mW, and this suggests that the Gaussian profile of the beam introduces significant spatial inhomogeneity in $T_1$ and $T_2$ at laser powers $\geq 6$ mW. As the microwave power is increased, we can expect that the full ensemble of NV$^-$ centers, inhomogeneously illuminated by the laser, will contribute to the dynamics, with each NV$^-$ center having a different transient response and fluorescence intensity. As an aid to visualizing the way in which the experimental curves include contributions from NV$^-$ centers that have a range of time constants, Fig. 2 shows representative experimental data, together with the simulated response of a single NV$^-$ center having a different time constant within the expected range at the corresponding laser power of 39 mW ($T_1=0.5 \mu s$, $T_2=0.1 \mu s$ and $T_1=15 \mu s$, $T_2=1.5 \mu s$) [SI].

In Fig. 3, we illustrate similar experiments repeated at various optical and microwave excitation powers, along with estimated bandwidths, defined as the modulation rate at which the amplitude decreases by 3 dB. At any given optical excitation power, the bandwidth increases monotonically with the microwave excitation powers, but with a larger absolute change at higher optical excitation power. For example, at an optical excitation power of 46 mW, measured before the microscope objective, the measured bandwidth increased to $\sim 1.6$ MHz as the Rabi frequency, $\omega_\delta$, was increased to 4.10 MHz, but at a lower optical excitation power of 0.45 mW, the bandwidth increased only to $\sim 39$ kHz when $\omega_\delta$ was increased over the same range.

The sensitivity of the ensemble diamond magnetometer to an oscillating magnetic field was compared for a single fluorescence channel and a gradiometer with two fluorescence channels. Microwave excitation was applied at $f_{MW}$, and a calibrated magnetic field from a thin wire was applied with a modulation rate of 2 kHz. The modulated NV fluorescence signal was integrated for 1 sec using a spectrum analyzer (Stanford Research 770) with a uniform window function.

As shown in Fig. 4, we measured a magnetic field sensitivity of $\sim 4.6$ nT/Hz using the gradiometer and $\sim 6.7$ nT/Hz using the single input channel. In the absence of technical noise, the sensitivity of such a magnetometer configured as a gradiometer (Fig. 1) is frequently limited by the photon shot noise [7], derived in the Supporting Information:

$$\delta B = \frac{4}{3} \frac{\Delta \omega}{\gamma R} \sqrt{\frac{1}{N \cdot T_c}},$$

Here, $\gamma$ is the electron gyromagnetic ratio, $\Delta \omega$ is the peak-to-
peak linewidth of the first derivative of the absorption spectrum, $R$ is the contrast, $N$ is the number of detected photons per unit time in the fluorescence signal, and $t_{\text{int}}$ is the signal integration time. Under our experimental conditions with pump power of 40.3 mW($\Delta \omega / 2\pi = 7.5 \pm 0.1$ MHz, $R \sim 0.043$, and detected fluorescence power $\sim 1.0 \mu$W), this yields a photon shot-noise limited sensitivity of $\leq 4.4$ nT for 1 s integration time, approximately the same as our measured sensitivity. Further sensitivity improvements will therefore require reducing the shot-noise limit and can be expected by increasing the signal contrast using polarization-selective NV excitation [10], by improving either the detector volume or the collection efficiency [11], or by using a diamond with more favorable properties, such as longer coherence times or a higher NV$^-$:NV$^+$ ratio.

If the sensitivity is limited by the photon shot noise, then it should also depend on the optical excitation power as follows:

$$\delta B_p \propto \frac{T}{N} \propto \sqrt{\frac{P_{\text{sat}}}{P}}, \tag{4}$$

where $P_{\text{sat}}$ is a characteristic saturation power of the NV defects, and $P$ is the applied optical excitation power [12]. The sensitivity of the NV$^-$ ensemble to a 2kHz oscillating magnetic field, was measured at various optical excitation powers from 2 to 45 mW and showed good agreement (inset in Fig. 4) with Eq. (4).

For its use in NMR, the magnetometer must detect weak, oscillating or modulated magnetic fields. To simulate this application, we employed a double-modulation technique in which we applied a frequency-modulated microwave excitation, centered at the zero crossing of the spectrum. The modulation amplitude was 4.5 MHz with modulation rates ranging from 10 to 100 kHz; we also applied an additional oscillating magnetic field of 11.9 nT/Hz using a gradiometer with this technique, limited by the additional electronic noise from the lock-in amplifier output that could not be removed with our present experimental apparatus.

In summary, we have developed an ensemble diamond magnetometer for NMR applications with a sensitivity of 4.6 nT/Hz, exceeding by a factor $\sim 5$ the best reported sensitivity for an ensemble NV$^-$ magnetometer operating at room temperature. We measured a detection bandwidth of 1.6 MHz and developed an analytical theory to explain the transient magnetic field response of an ensemble of NV$^-$ centers under continuous microwave and optical irradiation. Our results demonstrate that magnetic sensitivity of a few nT at room temperature can be achieved with ensemble NV$^-$ centers of short $T_2^*$ $\sim 130$ ns, using modulation techniques that make the measurements robust against most of the experimental noise. The results are relevant for the development of affordable, integrated, and portable diamond magnetometers for a variety of field-sensing applications.

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