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A differential photon-rate meter for real-time peak tracking in optically detected magnetic resonance at low photon-count rates

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Abstract

We describe a real-time method to obtain normalized differential count rate signals from modulated systems with photon detection at single-photon count rates. The method is demonstrated with a real-time peak-locking and frequency control to track magnetic field using optically detected magnetic resonance of nitrogen-vacancy centers in diamond. This procedure allows us to measure magnetic field continuously with sensitivity $4.1 \mu\text{T/Hz}^{1/2}$ and to track magnetic field sweep rates up to $50 \mu\text{T/s}$. The differential rate meter automates the processing of voltage pulse outputs from the photon detector, and provides noise levels on par with traditional photon counting methods using digital counters.

Introduction:

The work described in this paper was inspired by the quantum measurement capabilities of nitrogen-vacancy (NV) centers in diamond. At the core of these capabilities is an optically detected magnetic resonance, where the spin state of the NV center is measured via spin-dependent photoluminescence, and the long coherence time of the NV center's spin state allows precise magnetic field measurements [1] [2] [3]. NV centers also have demonstrated sensitivity to temperature [4][5][6][7], strain [8], and electric field [9], all using measurement schemes based on the respective effects on the magnetic resonance. Magnetic field measurements with noise floors below $1 \text{ nT/Hz}^{1/2}$ [10][11][12][13][14][15] have been demonstrated with very large ensembles of NV centers.

As a sensor, the NV center has an additional advantage of being small. Of course, the NV center itself is an atomic-scale object, but for sensing external objects the size of the diamond host crystal is the relevant length scale. The NV center must be formed at a depth

Contributions:

K.A. designed and performed the experiment, measurement analysis and contributed to the manuscript preparation. R.D.M and K.A. conceived the concept. R.D.M oversaw the project including analysis, discussion and conceptual guidance and he contributed to the manuscript preparation.

Competing interests:

Based on the real-time magnetic resonance peak tracking concept using ratemeter discussed in this study, an invention disclosure was made at the technology commercialization office of the University of Maryland.

on the order of 10 nm from the diamond surface and at a density needed to preserve its vital intrinsic properties like long coherence time, long relaxation time and good stability [16]. For any particular application, there are tradeoffs to be considered between the higher sensitivity and larger sensor size of very large ensemble measurements on one hand with reduced sensitivity and smaller sensor size of small ensemble including single-NV measurements on the other.

Real-time and precise measurement of magnetic field is important for many scientific and technological applications ranging from physics to biology [17,18]. As an alternative to measuring and fitting complete spectra, magnetic resonance peak locking and tracking methods provide for real-time and continuous measurements of the magnetic field. These methods include modulation of the applied field or the excitation frequency near resonance, producing a corresponding modulation of the NV⁻ centers photoluminescence. Near a resonance extremum, the modulation of the photoluminescence signal produces an “error signal” proportional to the deviation from resonance. This error signal is used in a feedback loop to adjust the excitation frequency to “lock” the resonance. The feedback-controlled excitation frequency value provides a real-time field measurement by effectively concentrating all measurements near the resonance extremum.

Such active feedback control for real-time magnetometry has been demonstrated [10][19][11] using large ensembles of NV⁻ centers, which emit relatively high photoluminescence power detected by regular photo-diodes and demodulated using lockin amplifiers. But for sensing using single NV⁻ centers or small ensembles of NV⁻ centers, their low photoluminescence power requires single photon detection, and the peak tracking method suffers from a technical problem. The collected photoluminescence from a single NV center yields photon count rates on the order of 10^5 s^{-1} , where single photon detectors (avalanche photo-diodes, APD) are required.

The discrete voltage pulses generated by APD are not amenable to demodulation by lockin amplifiers. The pulses are very narrow ($\approx 20 \text{ ns}$), producing a broadband spectrum. Additionally, the pulse signals place great demands on a lockin’s dynamic range. The lockin must be able to measure the pulse maxima while maintaining sufficient precision to resolve the small modulation of the time-averaged signal representing the photon detection rate. Instead of using lockin amplifiers, the conventional approach is to count pulses and communicate the results to a computer for demodulation and feedback control via computer algorithms [20] [21].

Here, we address active feedback control and real-time field tracking with photo detection rates in the range of $4 \times 10^3 \text{ s}^{-1}$ to $1000 \times 10^3 \text{ s}^{-1}$ using a custom differential rate meter with phase sensitive detection that generates a normalized difference signal from APD pulse inputs. Without compromising signal-to-noise, this real-time data processing scheme provides all the typical functionalities of a lock-in amplifier needed for real-time peak locking and tracking. In the experimental procedure section below, we describe our test setup. Following that, the Results and Discussion section presents an evaluation of the signal-to-noise ratio, sensitivity ($4.1 \mu\text{T/Hz}^{1/2}$), and real-time field tracking for sweep rates up to $50 \mu\text{T/s}$.

Experimental Procedure:

The experimental setup consists of three parts; 1) confocal microscope for optically detected magnetic resonance (ODMR) measurement using NV^- centers, 2) differential rate meter, and 3) frequency locking using a proportional-integral (PI) controller.

The confocal microscope for ODMR measurement consists of 1) a green laser for excitation, 2) a custom-built electromagnet for the applied magnetic field, 3) a microwave antenna to drive spin resonance, and 4) microscope objective with numerical aperture 0.55. The diamond sample used in the measurement was type-1b CVD-grown diamond crystal with <1 ppm nitrogen concentration which produces an estimated about <30 NV^- centers in the focal spot with axes aligned with the applied magnetic field. The NV^- centers are continuously excited with green laser light (532 nm) with nominal illumination range ≈ 100 μW to ≈ 300 μW , which is low enough to avoid optical power related broadening. Emitted photoluminescence (600 nm to 800 nm) is collected and detected by an avalanche photo diode (APD). The APD outputs a 20 ns, 5 V voltage pulse per detected photon. For ODMR measurement, the photon detection rate is then recorded using the rate meter as a function of microwave frequencies.

The rate meter demodulates the photoluminescence signal from the APD by switching the input pulse train between the inputs of two parallel rate-meter channels synchronously with the modulation. The overall effect of each channel is to integrate the incoming pulse train. Direct integration of the ≈ 20 ns long pulses at typical 10^4 s^{-1} rate produces a weak signal that is sensitive to variations in the pulse duration. To boost the signal and to reduce sensitivity to pulse duration, we stretch the pulses in each channel to uniform 200 ns duration by triggering monostable multivibrator circuits. With this step, we increase the effective dead time, which limits the maximum count rate to $\approx 10^8$ s^{-1} . Integration is performed in a frequency-to-voltage circuit based on a voltage-controlled oscillator with negative feedback regulation. These integrators function at pulse rates from 2×10^3 s^{-1} to 500×10^3 s^{-1} . Finally, the normalized difference is obtained from the individual channel voltages using an analog multiplier/divider chip and low-pass filtered with time constants ranging from 100 μs to 1 s.

The rate meter is used with either amplitude modulation or frequency modulation. For amplitude modulated (AM) cw-ODMR measurements, the microwave power alternates between ON and OFF states. In the OFF state, the NV^- centers are continuously pumped into the bright $|0\rangle$ spin state, while in the ON state, photoluminescence is reduced as spins are driven into $| - 1 \rangle$ and $| + 1 \rangle$ states. The normalized differential output $(A-B)/(A+B)$ is recorded as a function of the microwave frequency. Results are shown in Figure 2.

For frequency modulated (FM) cw-ODMR measurements, which we use for peak tracking, the microwave frequency alternates between two frequencies, f_1 and f_2 , around a center frequency, f_c . Channel-A is open, receiving pulses, while the frequency is f_1 and channel-B is open while the frequency is f_2 respectively. Similarly, to the amplitude-modulated case, we record the frequency-modulated, normalized-difference, cw-ODMR spectrum as a function of microwave frequencies (f_c) as shown in figure-3a.

The derivative-like line shape of the frequency-modulated signal provides an error signal for locking the resonance peak. Near resonance, the demodulated signal is positive if the driving frequency is above resonance, and negative if the driving frequency is below resonance. We use active feedback to generate a frequency-correction signal that adjusts the microwave frequency. We find that digitizing the correction signal and using computer control of the microwave generator frequency yields good performance. The slope of the frequency modulated signal is used as a voltage-to-frequency conversion factor.

Results and discussion:

First, we compare results obtained using the rate meter with results from the more conventional method of direct pulse counting. We measured amplitude modulated cw-ODMR spectra at a fixed applied magnetic field and recorded data for the two methods simultaneously, using copies of the same pulse train. In this measurement, the microwave power was modulated at 50 Hz: ON for 10 ms and OFF for the subsequent 10 ms in each cycle. The average emitted photon count rate was $300 \times 10^3 \text{ s}^{-1}$ and average incident laser power was 112 μW . ODMR spectra from both measurements were normalized, as shown in Figure 2. The blue, square points are from the conventional photon counting method and the black, circular points are from the rate meter method. As shown in Figure 2, the signal-to-noise ratios (SNR) from the two methods are virtually identical.

We now turn to using the field-dependence of the ODMR signal for magnetometry. The noise floor of a field measurement, δB , can be determined from the signal noise, δS_{FM} , of a frequency-modulated measurement using

$$\delta B = \frac{dB}{df} \left(\frac{dS_{\text{FM}}}{df_c} \right)^{-1} \delta S_{\text{FM}}.$$

The differential sensitivity, $\frac{dS_{\text{FM}}}{df_c}$, is the slope of the frequency modulated cw-ODMR

spectrum as shown in Figure 3a. In this measurement, microwaves were frequency-modulated using a square wave. The modulation rate was 500 Hz and the amplitude was 3 MHz. Channel-A received all the photons when the microwave frequency was high, ($f_1 = f_c + 3 \text{ MHz}$) and channel-B received all the photons when the microwave frequency was low ($f_2 = f_c - 3 \text{ MHz}$). The normalized difference was measured as function of the center microwave frequency (f_c) for an integration time of 1 s per point. The noise level was determined from the off-resonance background, as shown in the dotted boxes. The histogram of the background signal is shown in Figure 3b; the standard deviation is $\delta S_{\text{FM}} = 0.69 \text{ mV}$ with a bandwidth of 1 Hz. We also determined the frequency sensitivity from a linear fit of the data points around the zero-crossing. The fit is shown as a red-line in Figure 3c. The measured slope $\frac{dS_{\text{FM}}}{df_c}$ for this fit is $6.07 \times 10^{-6} \text{ mV/Hz}$. The measured frequency sensitivity was $\approx 114 \text{ kHz/Hz}^{1/2}$, which translates via the gyromagnetic ratio into a field sensitivity of $4.1 \mu\text{T/Hz}^{1/2}$.

Next, we demonstrate the peak tracking method for a wide-range magnetic field measurement. In this measurement, we lock and follow the magnetic resonance peaks of NV^- centers from near zero magnetic field to 35 mT, revealing the Zeeman splitting of the NV^- center as shown in Figure 4a. At magnetic fields near zero, the tracking feedback loop becomes unstable as the two resonance peaks overlap. The magnetic field (bottom axis of fig. 4a) was independently measured using a Hall probe placed near the sample. The result in figure-4a is in agreement with the expected Zeeman splitting, and also shows the linearity of the probe and the detection scheme over wide a magnetic field range. The intercepts from the linear fits are virtually identical: 2.868 GHz and 2.869 GHz in agreement with the expected zero field splitting of the NV^- center [22]. The uncertainty estimates from the standard deviations of the fit parameters are approximately 0.7 MHz, but systematic errors such as the field offset of our Hall probe may dominate. Figure 4b and 4c display the corresponding fit residual. Figure 4d is the portion of the upper plot (shown in dotted box) from 20 mT to 22 mT, showing individual data points.

In principle, the potential field range of this measurement scheme is determined by the frequency range of microwave components such as the microwave frequency source, RF cables and the microstrip antenna. In this demonstration, however, we are limited by the maximum field of our small, air-cooled electromagnet.

To demonstrate real-time data processing, frequency control, and field tracking of the feedback scheme, we applied an oscillating magnetic field of the form $B(t) = B_0 \cos(2\pi ft)$ with various frequencies f and amplitudes B_0 . In fig. 5a and fig. 5b, the applied oscillating magnetic field amplitude B_0 was 154 μT and frequencies were 0.1 Hz and 1 Hz correspondingly. The insets of figs. 5a and 5b are the time traces of the feedback output in real-time. The sampling time for this measurement was 20 ms. Fig. 5c compares the maximum magnetic field sweep rate, $2\pi f B_0$, measured by the real-time feedback scheme with the maximum sweep rate measured using the Hall probe. The x-axis of the fig. 5c is the maximum magnetic field sweep rate measured using Hall probe placed near the sample and y-axis of the fig. 5c is the rate of change detected by feedback loop. The square (green) data points of fig. 5c are measured by applying sinusoidal varying magnetic field of variable amplitude at a frequency 0.1 Hz. The circle (red), triangular (blue) and star (pink) data points are measured in frequency range 0.1 Hz to 3 Hz for a fixed amplitude. The oscillating magnetic field amplitude of these measurements were 154 μT , 231 μT and 308 μT respectively.

Fig. 5c shows that at lower rates the feedback system exactly follows the applied oscillating magnetic field but at rate higher than approximately 50 $\mu\text{T/s}$, the feedback scheme is unable to follow the change in magnetic field.

Conclusion:

In conclusion, we implemented a frequency modulated differential photon rate measurement for optically detected magnetic resonance of NV^- centers using single photon detectors. We demonstrated that the rate meter method provides virtually identical signal-to-noise ratios as compared to conventional photoncounting methods. Using differential measurement and

active feedback (PID) control, we lock and follow the magnetic resonance peak of NV⁻ centers from near-zero fields to 35 mT. The sensitivity and maximum tracked-field sweep rate of our detection scheme are 4.1 $\mu\text{T}/\text{Hz}$ and 50 $\mu\text{T}/\text{s}$ respectively. The implementation of our peak-tracking method is relatively simple and could easily be integrated with other experimental setups such as scanning probe microscopy for nanoscale magnetometry. Also, the described rate meter can detect photon rates from $4 \times 10^3 \text{ s}^{-1}$ to $1000 \times 10^3 \text{ s}^{-1}$. Therefore, this method is well suited for use in single NV⁻ center as well as small ensembles of NV⁻ centers.

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Fig.1.

Magnetic resonance tracking system using NV⁻ centers as magnetic field sensors. NV centers are excited with green laser light and emitted photoluminescence is collected using an avalanche photodiode (APD). The APD signal is sent to the differential rate meter, which contains two gated high-speed voltage pulse integrators, a low pass filter (LPF) and an analog operation for a normalized differential output. The PI controller monitors any changes in the output of the rate meter and generates a compensating frequency-control output that tracks the ODMR resonance peak.

Fig. 2.

Amplitude-modulated, cw-ODMR spectra measured using both the differential rate meter and the traditional counter method on identical input signals. Data sets are offset for better visibility. Black circles represent the data recorded using the rate meter and blue squares represent data recorded using pulse counters. Signal-to-noise ratios are virtually identical.

Fig. 3.

a) Frequency modulated cw-ODMR spectrum of NV- centers measured using the differential rate meter. Average incident laser power was $\approx 300 \mu\text{W}$ and average emitted photon counts rate was $800 \times 10^3 \text{ s}^{-1}$. The rate of frequency modulation was 500 Hz with an amplitude of 3 MHz. b) Histogram of the background noise taken from the data points shown as dotted box in fig.3a. c) A fit to the points nearest the zero crossing in a).

Fig4.

Continuous magnetic field measurement using custom-built rate meter and feedback control for peak tracking. Figure-4a shows the Zeeman effect of NV- centers, measured by tracking the resonance peaks. Figures 4b and 4c plot the residual of linear fits. Figure 4d is a magnified portion of the upper curve outlined by the dotted box from 20 mT to 22 mT.

Figure 5.

Real time data processing and frequency control using a rate meter and feedback control. (a) and (b) are the FFTs of time traces of V_{out} as shown in the respective inset. The oscillating magnetic field amplitude is $154 \mu T$ and the frequencies were 0.1 Hz and 1 Hz . (c) is the measured maximum field sweep rate plotted against the maximum sweep rate of the applied field.