Demonstration of vector magnetic field sensing by simultaneous control of nitrogen-vacancy centers in diamond using multi-frequency microwave pulses

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An ensemble of nitrogen-vacancy (NV) centers in diamond is a fascinating candidate to realize a sensitive magnetic field sensor. In particular, since the axes of the NV centers are distributed along four directions, a collection of measurement data from NV centers with different axes provides information on the vector components of a magnetic field. However, in the conventional approach, the low measurement contrast of NV centers limits the sensitivity of vector magnetic field sensing. Recently, to overcome this problem, multi-frequency control of the NV centers has been proposed. The key idea is that the four types of NV centers with different axes are simultaneously controlled by multi-frequency microwave pulses. Here, we demonstrate vector magnetic field sensing with an ensemble of NV centers in diamond via such multi-frequency control with pulsed-type measurements. We use Hahn echo pulses and extract information on the vector components of an applied AC magnetic field. We find that the sensitivity of diamond-based vector field sensing with multi-frequency control is better than that with single-frequency control for every vector component of a magnetic field.

Magnetic field sensors have significant applications in chemistry, biology, and medical science. For example, in electron spin resonance, which is a widely used technique in chemistry, magnetic field sensors play an important role in obtaining information about the electron spin. Magnetoencephalography is a clinical technique for measuring electrical activity in the brain via magnetic field sensing, thereby providing information about brain function. Magnetic resonance imaging allows examination of the internal structure of the human body based on magnetic field information. For the magnetic field sensors used in these contexts, sensitivity and spatial resolution are essential parameters to quantify performance, and much effort has been devoted to measuring weak magnetic fields in local regions.

Qubit-based sensors are attractive candidates for use in such applications. A solid-state qubit can be coupled with a magnetic field, resulting in a shift in the resonant frequency of the qubits. The associated energy shift can be detected via Ramsey measurements in the case of an applied DC magnetic field. It is also possible to detect an AC magnetic field by using Hahn echo pulse sequences, which suppress dephasing of the qubits, thereby typically improving sensitivity. A number of types of qubit sensor have been demonstrated experimentally, using, for example, atoms, superconducting flux qubits, or ion traps.

One important type of qubit-based sensor uses nitrogen-vacancy (NV) centers in diamond. The NV center is a spin-1 system, and magnetic fields can change the energies of the states |±1⟩. By using frequency selectivity, we can consider an NV center as an effective two-level system comprising |0⟩ and |1⟩, with |−1⟩ being energetically well detuned from the other states. Since NV centers can be strongly coupled with optical photons, it is possible to initialize and read out the spin state of |0⟩ and |1⟩ (|−1⟩) by applying a green-light laser. High-fidelity gate operations with NV centers have been demonstrated using microwave pulses. Long coherence times in the region of a few milliseconds have been observed even at room temperature. Owing to these properties, NV centers can be considered as suitable systems to use in sensitive magnetic field sensors.

Moreover, NV centers are useful for the measurement of the vector components of applied magnetic fields. As can be seen in Fig. 1(a), NV centers have four possible crystallographic axes: [111] (NV1), [111] (NV2), [111] (NV3), and [111] (NV4). The vector component of a magnetic field along the direction of one of these axes will induce a frequency shift in an NV center with that axis. Thus, a collection of the measurement data from the four types of NV center with different axes provides information on all the vector components of a magnetic field. There have been a number of experimental demonstrations of the use of NV centers to measure vector components of magnetic fields. For example, vector components of AC magnetic fields from current-carrying wires have been measured and analyzed using this technique, and vector imaging of DC magnetic fields from living cells and circuit currents has been demonstrated. Attempts have also been made to increase the signal-to-noise ratio of vector measurement by combined RF excitation of hyperfine triplets in the optically detected magnetic resonance (ODMR) spectrum.

However, such diamond-based sensors are affected by low readout contrast, which significantly decreases their sensitivity for vector fields. Although photoluminescence (PL) from the state |0⟩ is slightly larger than that from the state |1⟩, most of the photons emitted from the NV centers are absorbed in the environment, and so a large number of repetitions are typically needed to determine the population of the state |0⟩.
In this paper, we demonstrate vector magnetic field sensing with multi-frequency control. We adopt a spin echo measurement technique, which is one of the typical pulsed ODMR schemes for measuring AC magnetic fields using NV centers. In a spin echo measurement, by irradiating NV centers with the microwave pulse sequence shown in Fig. 1 (b), the AC magnetic field induces a phase shift on the state of the electron spin. By reading out the state using the green laser, we can detect the amplitude of the AC magnetic field. Moreover, in the conventional approach to vector field sensing, during measurement of any one of the four types of NV center, the other three types remain in the state $|0\rangle$ regardless of the applied magnetic field, which induces additional noise.

Recently, to overcome this problem of low readout contrast, an approach to vector magnetic field sensing has been proposed in which the four types of NV center with different axes are simultaneously controlled by multi-frequency microwave pulses. Consequently, all the NV centers irradiated by a green laser contribute to the signal containing information on the magnetic field, in contrast to the conventional approach, in which 75% of the NV centers just induce noise without contributing to the signal. The sensitivity of this method of vector field sensing can, in principle, be four times higher than that of the conventional approach. It is worth mentioning that vector magnetic field sensing with simultaneous control of NV centers has been achieved using CW-ODMR measurements, but multi-frequency control using pulsed-type measurements (which typically have the advantage that they can suppress dephasing) has not yet been demonstrated.

![Experimental setup. Microwaves (MW) are generated by two sources, and are modulated by plus and minus sidebands at radiofrequency from a function generator to produce multiple frequencies for the control of NV centers with four different axes. They are amplified, combined, and radiated by a MW antenna. MW pulses are generated by four switches, which are controlled by a pulse blaster. The excitation laser beam passes through an acousto-optic modulator (AOM), which is used to switch the laser. After the AOM, the beam passes through an optical fiber and objective lens, whose position is controlled by a piezo stage. PL from the NV center passes through a confocal system and long-pass filter at 630 nm and is collected by an avalanche photodiode (APD). The target AC magnetic field is generated by a copper wire.](image)

FIG. 1. (a) The structure of an NV center in diamond. This contains a nitrogen atom (substituting for a carbon atom) and a vacancy in the diamond lattice sites. NV centers have four possible crystallographic axes. (b) Microwave pulse sequence for multi-frequency control of NV centers. In this case, the phase of the last microwave pulse is shifted by $180^\circ$. For NV2 and NV4, the phase shift of the microwave pulse is required for the x component, the same phase shift of the microwave pulse is required for NV3 and NV4 (NV2 and NV3).

FIG. 2. Experimental setup. Microwaves (MW) are generated by two sources, and are modulated by plus and minus sidebands at radiofrequency from a function generator to produce multiple frequencies for the control of NV centers with four different axes. They are amplified, combined, and radiated by a MW antenna. MW pulses are generated by four switches, which are controlled by a pulse blaster. The excitation laser beam passes through an acousto-optic modulator (AOM), which is used to switch the laser. After the AOM, the beam passes through an optical fiber and objective lens, whose position is controlled by a piezo stage. PL from the NV center passes through a confocal system and long-pass filter at 630 nm and is collected by an avalanche photodiode (APD). The target AC magnetic field is generated by a copper wire.
Figure 2 shows our experimental setup for multi-frequency control vector magnetic field sensing. We use a home-built system for confocal laser scanning microscopy with a spatial resolution of 400 nm. The diamond sample is positioned above the antenna used to generate the microwaves. A magnet is placed below the antenna to apply a static magnetic field, and a copper wire is placed in contact with the sample surface to apply the target AC magnetic field. We use a single-photon-resolving detector to measure the photons from the NV centers. Microwaves of four different frequencies are generated by modulating the microwaves from two sources, each of which gives two frequencies, using in-phase/quadrature (IQ) mixers and function generators. The microwave phase “x” is controlled by switches 1 and 3 (LO + I) and the phase “y” by switches 2 and 4 (LO + Q).

The details of the diamond sample used in our experiment are as follows. We use an ensemble of NV centers in a 2 \( \mu \)m-thick diamond film on a (001) electronic-grade substrate. Diamond films were prepared from an isotopically-enriched (> 99.999% for \(^{12}\)C) \(^{12}\)CH\(_4\), H\(_2\) mixed gas system in a microwave plasma-assisted chemical vapor deposition. To both increase the NV center density and improve the coherence time, the sample was irradiated with 350 keV He\(^{+}\) ions at ion doses of \(10^{12}\) cm\(^{-2}\), followed by annealing for 24 hours in vacuum at 800 °C. To ensure a uniformly random distribution about the four possible crystallographic axes, with each orientation rate being around 25%, the sample was annealed for 15 hours in vacuum at 1200 °C. The NV center density of this diamond sample is \(2.2 \times 10^{15}\) cm\(^{-3}\) and the coherence time is around \(20 \mu s\).

We demonstrate vector AC magnetic field sensing with a frequency of 100 kHz by multi-frequency control. First, we measure the ODMR spectrum and Rabi oscillations, as shown in Fig. 3. An applied static magnetic field separates the frequencies of NV centers with the four different axes, and we adjust the Rabi frequency around 2.5 MHz (which corresponds to a time scale for the \(\pi\) pulse of around 200 ns). From the ODMR spectrum, we determine the resonant frequencies of the NV centers with different axes: \(f_1 = 2.72\) GHz (NV1 = [111]), \(f_2 = 2.806\) GHz (NV2 = [111]), \(f_3 = 2.826\) GHz (NV3 = [111]), and \(f_4 = 2.862\) GHz (NV4 = [111]). From the behavior of the Rabi oscillations, we determine
the orientation ratios as (NV1) : (NV2) : (NV3) : (NV4) = 29% : 35% : 21% : 15%.

Second, we consider a particular case and measure the amplitude of an AC magnetic field under the assumption that its direction is known. To find the sensitivity of the conventional scheme, we measure the optical signal against the magnetic field amplitude and also measure the signal fluctuation against the repetition time when only a single-frequency microwave is used to control the NV centers. These results are shown in Fig. 4 (f1, . . . , f4). The uncertainty of the estimation is given by \( \delta B_n = \delta P_n/(dP_n/dB_n) \) (n = f1, . . . , 4, multi), and we obtain the sensitivity of each axis from Fig. 4 as follows (units of nT/Hz): 80 ± 11 (NV1), 76 ± 8 (NV2), 85 ± 11 (NV3), and 85 ± 8 (NV4). We then perform similar measurements to determine the sensitivity of our approach with multi-frequency control of the NV centers. Here, to enhance the optical signal, we shift the phase of the last microwave pulse by 180° for f1 and f4 (NV1 and NV4), so that the sign of the signals from NV1 and NV4 around 0 µT becomes the same as the sign of those from NV2 and NV4. These results are also shown in Fig. 4. We obtain the sensitivity of measurement of the AC magnetic field with our scheme as \( \delta B_{\text{inf}} = 35 ± 5 \text{nT/\sqrt{Hz}} \), which is approximately twice as good as the sensitivity of the conventional scheme.

Finally, we measure the vector components of the AC magnetic field under the assumption that we do not know the direction of the field. In Fig. 5 we plot the echo intensity against the amplitude of the AC magnetic field for three different pulse sequences corresponding to the extraction of the three vector components \( B_x, B_y, \) and \( B_z \), respectively. Using our multi-frequency control scheme, we obtain \( (B_x, B_y, B_z) = (0.23, 0.16, -0.97) \), where the tilde \(~\) denotes the dimensionless normalized vector component. For comparison, we also measure the vector components using conventional single-frequency control and obtain nearly the same result: \( (B_x, B_y, B_z) = (0.23, 0.17, -0.96) \). This demonstrates the ability of our multi-frequency control method to measure each vector component of an applied magnetic fields. Also, we compare the sensitivities of measurement of \( B_x, B_y, \) and \( B_z \) between our scheme and the conventional one. We define the uncertainties in the estimation of the vector components using the conventional scheme and our multi-frequency scheme as

\[
\delta B_x^{(c)} = \frac{\sqrt{2} \delta P_{\text{single}}}{(dP_{(111)} - dP_{(111)})/dB_x},
\]

\[
\delta B_x^{(mf)} = \frac{\delta P_{\text{multi}}}{dB_x},
\]

where \( P \) denotes the optical signals from the NV centers. We define the sensitivities for the \( y \) and \( z \) components similarly. It is worth mentioning that to measure one of the vector components of the magnetic field using the conventional method, measurements of two signals from NV1 (||111||) and NV3 (||111||) are required, which doubles the measurement time. This is the reason for the factor \( \sqrt{2} \) in the numerator of the sensitivity for the conventional scheme in Eq. (1). From Fig. 4 (f1, . . . , f4), in the conventional scheme, the vector sensitivities are obtained as as follows (units of nT/√Hz): \( \delta B_x^{(c)} = 120 ± 30, \delta B_y^{(c)} = 200 ± 60, \) and \( \delta B_z^{(c)} = 41 ± 6 \). In our scheme, the vector sensitivities are obtained from Fig. 5 as follows (units of nT/√Hz): \( \delta B_x^{(mf)} = 35 ± 10, \delta B_y^{(mf)} = 42 ± 16, \) and \( \delta B_z^{(mf)} = 34 ± 5\). Therefore, we conclude that our scheme shows better sensitivity than the conventional scheme for every vector component of the AC magnetic field. In these demonstrations, \( \delta B_z^{(mf)} \) is close to \( \delta B_{\text{inf}} \). When we measure \( \delta B_{\text{inf}} \), the magnetic field direction is near to the \( z \) direction, and so these experimental results are consistent in our setup.

The theoretical ratios between the sensitivities of our scheme and those of the conventional scheme for each vector magnetic field component can be calculated as follows. The ratio of the sensitivities for \( x \) and \( y \) components is

\[
\frac{\delta B_x^{(c)}/\delta B_x^{(mf)}}{\delta B_y^{(c)}/\delta B_y^{(mf)}} = \frac{\delta B_x^{(c)}}{\delta B_y^{(c)}} \approx 4,
\]

and thus the ratio should be the same for each magnetic field component. However, in our experiment, the sensitivity improvement ratio depends on which vector component we have extracted for the vector field sensing. One possible reason is imperfect application of the microwave pulses with four frequencies in our setup. The use of a microwave antenna to recover the sensitivity could solve this problem. The discrepancy could also be due to the fact that the ratios of the crystallographic orientations are not equal among the four directions in our experiment, whereas the theoretical calculations assume a 25% orientation ratio for all four directions. Such an inhomogeneous distribution of orientations means that the sensitivity depends on which vector component is measured in our scheme. In principle, we could overcome this problem by more sophisticated sample fabrication. Moreover, adjusting the pulse sequence period could, in principle, change the effective orientation ratio.

In conclusion, we have demonstrated vector magnetic field sensing with multi-frequency control of NV centers in diamond. We have achieved a sensitivity twice better than that of the conventional scheme. By choosing an appropriate pulse sequence, we can extract the vector components of a magnetic field. Although we use a spin echo method in our demonstration in this paper, the technique described here is quite general.
and could be applied with other methods, such as DC magnetic field sensing with Ramsey measurement or AC magnetic field sensing with dynamical decoupling for further improvement in sensitivity.

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