Dressed-state Generation for Virtual Memories toward Hybrid Quantum Sensing

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Paramagnetic centres in a solid hold promise in future sensing applications. Numerous sensing applications have been theoretically and experimentally demonstrated. However, sensitivity improvements remain challenging. One approach to overcome this is hybrid quantum sensing with quantum memories. We propose hybrid quantum sensing with virtual quantum memories using dressed states. We also observe the generation of more than four dressed states in a single paramagnetic centre based on Autler-Townes splitting (ATS) and demonstrate the extension of the coherence time to $T_2 \approx 1.5$ ms for dressed states, which is more than two orders of magnitude longer than that of the undressed spin. Furthermore, we theoretically estimate the enhancement of sensitivity by more than an order of magnitude compared with that without memory. The results and our approach will pave the way for new hybrid quantum sensing.

Nitrogen-vacancy (NV) centres in diamond, which have single spin manipulations and fluorescence measurements of the spins with a long coherence time ($T_2$) under ambient conditions, are promising candidates for classical- and quantum-sensing applications. Numerous experimental demonstrations have been performed such as sensing for temperature, imaging of living cells to understand drug-delivery systems, and atomic-scale structure analyses. To date, various approaches to improve the sensitivity of NV based sensors have been experimentally demonstrated. Hybrid quantum sensing, which is inspired by quantum memory effects, is one candidate.

We consider hybrid quantum sensing using the electron spin of an NV centre as a sensor and the nuclear spins of $^{13}$C or $^{14}$N around the NV centre as quantum memories. If paramagnetic impurities such as substitutional nitrogen (P1) centres are sufficiently suppressed and the interaction between the electron spin and the nuclear spin is weak, we can keep $T_2$ long. In such a case, a higher sensitivity can be achieved. However, when we considered situations to increase the number of nearby nuclear spins to develop our previous hybrid quantum sensor, $T_2$ of both the electron and nuclear spins became shorter. Consequently, the sensitivity decreased due to the trade-off between the number of nuclear spins and $T_2$.

Here we theoretically propose virtual quantum memories by dressed states generated on one nitrogen nuclear spin in an NV centre instead of using the real nuclear spins of $^{13}$C or $^{14}$N around the NV centre as quantum memories. Figure depicts our proposal toward hybrid quantum sensing using dressed states. Autler-Townes splitting (ATS) generates dressed states in an NV centre by driving microwave (mw) fields. Then we can manipulate the number of the dressed states by adjusting the intensity of the mw fields. It has been also reported that $T_2$ increases when dressed states are generated. Using these facts, we can realise a higher sensitivity by manipulating the number of dressed state as virtual quantum memories with an extension of $T_2$. First, we observe experimental generation of dressed states in a single NV centre in diamond by ATS at ambient conditions in order to analyse fundamental phenomena. Then we show the experimental result of an extension of $T_2$ and a theoretical estimation of the sensitivity as functions of the number of the dressed states and $T_2$ as an example of AC sensing.

In our experiment, dressed states are generated using an electron spin and the $^{14}$N nuclear spin of the single NV centre in diamond by irradiation of an mw drive field (Fig. 1(a)). In Fig. 1(a), $|m_s, m_f\rangle$ is defined as the electron and the $^{14}$N nuclear spin of the NV centre. After laser illumination, the population is in the $|0, 0\rangle$, $|0, 1\rangle$, or $|0, -1\rangle$ under $B_0$, which is depicted by the open circles.
THEORETICAL MECHANISM OF AUTLER-TOWNES SPLITTING

We briefly explain the mechanism for a dressed-state generation by ATS \cite{17, 22}. Figure 1 shows an unperturbed drive field, which has a nearly resonant frequency between $|0, 1\rangle$ and $|-1, 1\rangle$. When the NV centre is coupled to the mode of the drive field, which has a few energy levels (called a weak drive field), each $|0, 1\rangle$ and $|-1, 1\rangle$ is split into two levels described in Fig. 1(c). Here we focus on the generation of four dressed states of $|0, 1\rangle\{1\}$, $|1, 1\rangle\{0\}$, $|0, 1\rangle\{2\}$, and $|1, 1\rangle\{1\}$ in the presence of coupling between the NV centre ($|0, 1\rangle$ and $|-1, 1\rangle$) and the mode of the drive field ($|0\rangle$, $|1\rangle$, and $|2\rangle$), depicted in Fig. 1(c). This phenomenon is called (weak) ATS. It should be noted that Fig. 1(c) depicts the minimum number of dressed states by the ATS as an example. In the discussion on the sensitivity of our sensing strategy, we refer to the increase in the number of dressed states by the power of the drive field. Figure 1(c) also shows the dressed energy levels are characterised by the Rabi frequency of an NV electron spin ($\Omega$) and driving frequency ($\nu$), and its spectrum is given by the following equation \cite{22}:

$$f(\nu) = 2\pi \left( \frac{(\Delta \omega)^2 + \frac{1}{4}\kappa^2}{\Omega^2} \right) \delta(\nu - \omega) + \frac{\frac{1}{4}\kappa}{(\nu - \omega)^2 + \frac{1}{4}\kappa^2} + \frac{\frac{1}{4}\kappa}{(\nu - \omega - \Omega)^2 + \frac{9}{16}\kappa^2} + \frac{\frac{1}{4}\kappa}{(\nu - \omega + \Omega)^2 + \frac{9}{16}\kappa^2},$$

where $\nu$ is the incident probe-frequency, $\kappa$ is the inverse of the dephasing time, $\Delta \omega = \omega_0 - \omega$. Here $\omega_0$ means the resonant frequency of an NV electron spin.

The form of Eq. (1) shows that a single peak is split into three peaks under an unperturbed weak drive field. Their splitting widths describe the function of $\Omega$, which satisfies the following relation \cite{23}: $\Omega = \sqrt{\Omega_0^2 + (\Delta \omega)^2}$, where $\Omega_0$ is the Rabi frequency in the on-resonance condition. When $\Delta \omega = 0$, the splitting width $\Omega$ is equal to $\Omega_0$. It shows that the dressed-state position has linear dependences on the strength of the drive field ($B_{\text{drive}}$). On the other hand, when $\Delta \omega \neq 0$, the dressed-state position has two dependencies under the same strength of the drive field. One is a linear dependence on $\Omega$. The change in the central peak, which is described by the second term in Eq. (1), depends on $\Delta \omega$. Whilst, the other is a nonlinear dependence of $\Omega$. Note that the changes of the side peaks, which are described by the third and fourth terms in Eq. (1), originate from the following relation under the ATS \cite{23}: $\Omega = \Omega_0 \pm \sqrt{\Omega_0^2 + (\Delta \omega)^2}$. 

FIG. 2. (a) Schematic of a homemade confocal microscope with an electromagnetic field (emf) irradiation system. (b) Photoluminescence scanning image of the NV centres in diamond. Red circle shows the single NV centre used in this experiment. (c) \(g^{(2)}(\tau)\) for the NV centre. (d) Pulse sequence to demonstrate ATS.

EXPERIMENTAL GENERATION OF DRESSED STATE BY ATS

In our experiment, we chose an NV centre that is weakly coupled to other nuclear spins (e.g., \(^{13}\)C). The sample is a high-temperature high-pressure (HTHP) type IIa (111) diamond. Our experimental setup is shown in Fig. 2(a). (See Methods.) The second-order autocorrelation function, \(g^{(2)}(\tau)\), using the Hanbury-Brown-Twiss (HBT) setup \cite{24} was measured to confirm a single NV centre in the circle in Fig. 2(b). The power of the green laser is 100 \(\mu\)W. The measured \(g^{(2)}(0)\) is \(\sim 0.1\) (Fig. 2(c)). Therefore, this NV centre is a single centre. We also measured the optically detected magnetic resonance (ODMR) spectrum with a 1-\(\mu\)s laser pulse by sweeping the probe frequency of a 5.5-\(\mu\)s probe pulse (\(\pi\) pulse). In Fig. 2(a), the ODMR spectrum has three dips with 2.1 MHz splittings. These correspond to the hyperfine splitting of the \(^{14}\)N nuclear spin of the NV centre \cite{22}. This indicates that hyperfine couplings with other nuclear spins are weaker than ones with the \(^{14}\)N nuclear spin.

First, we measured the shifts of the dressed-state positions by changing the drive power for the three driving frequencies to observe the generated dressed states, 2834.75 MHz (Drive 1), 2837.05 MHz (Drive 2), and 2839.18 MHz (Drive 3), with the pulse sequence shown.
FIG. 3. (a) ODMR spectrum without any drive fields. (b) Resonant frequencies as a function of the strength of the drive field ($B_{\text{drive}}$). Black, red, and blue plots show the resonant-frequency shift by driving frequencies of 2834.75 MHz (Drive 1), 2837.05 MHz (Drive 2), and 2839.18 MHz (Drive 3), respectively. Solid lines are fitted for each resonant frequency. We can observe the Mollow triplet, which we call ATS. (c) ODMR spectrum under a drive field at a frequency of 2834.75 MHz and a drive power of 33 $\mu$T.

Next, we measured the shifts of the dressed-state positions under $\Delta \omega \neq 0$ by changing the driving frequency while fixing the drive power at 33 $\mu$T with the pulse sequence shown in Fig. 2(d). The driving frequency was changed by the step of 0.2 MHz. The result is shown in Fig. 4(b). When the driving frequency is close to a resonant frequency, the position is shifted from each resonant frequency. These results imply that the drive field is affected by the two energy levels whose energy difference is close to the energy of the drive field.

To understand more details of the off-resonant drive field impact, Fig. 4(b) plots the dressed-state positions as a function of the driving frequency around the centre of 2835 MHz. The black square in Fig. 4(b) shows the position shift with a driving frequency of $\sim$ 2835 MHz. The solid black line shows the linear fitting line. These results agree well with the theoretical prediction of $\Omega = \sqrt{\Omega_0^2 + (\Delta \omega)^2}$, where $\Omega_0$ describes the energy of the drive field. The red circle and the blue triangle in Fig. 4(b) also show the positions as functions of the driving frequency. They have non-linear dependences. This is because the shifts originate from Eq. 1 by the relation of the $\Omega = \Omega_0 \pm \sqrt{\Omega_0^2 + (\Delta \omega)^2}$. Thus, all the results are satisfied with the theoretical prediction in ATS, demonstrating that we successfully generate more than four dressed states under the ATS.
ESTIMATION OF THE SENSITIVITY OF OUR HYBRID QUANTUM SENSING

We discuss the sensitivity of our hybrid quantum sensing with virtual quantum memories based on dressed states. The generation of the dressed states gives us our model two effects, a 'memory effect' and an 'extension of $T_2$.' To obtain a higher sensitivity, we have to prepare more dressed states. Thus, we need to boost the strength of the mw-driving field such as more than tenfold of the mw-driving field $|2\hbar\Omega|/\hbar^2$.

Here we shortly explain how to generate an arbitrary number of the dressed states. We assume an NV centre is coupled with a mode of a strong mw-driving field, as illustrated in Fig. 5. The first energy is $\hbar\omega/2$, and the other states are separated by $\hbar\omega$, where $\hbar$ is the reduced Planck constant. If such a mode of the driving field of $|n\rangle$ is coupled with two states of the NV centre of $|1\rangle$ and $|2\rangle$, new dressed states $|D(n)\rangle_\pm$ appear. The $|D(n)\rangle_\pm$ and its energies $E_\pm(n)$ are described by the following equations:

$$|D(n)\rangle_\pm = c_1 |1, n\rangle \pm c_2 |2, n-1\rangle,$$

$$E_\pm(n) = \left( n - \frac{1}{2} \right) \hbar\omega \pm \frac{1}{2} \hbar\Omega,$$

respectively. $c_1$ and $c_2$ are the coefficients, which satisfy $|c_1|^2 + |c_2|^2 = 1$. In such a process, the number of the dressed states ($\geq 4$) can be increased by increasing the driving power, as depicted in Fig. 5.

Using this mechanism to manipulate the number of dressed states generated, we discuss the sensitivity of our proposed hybrid quantum sensing. Here it should be noted that AC magnetic field sensing is considered in Ref. 4. The key of our sensing is to integrate the information of the AC magnetic field in the virtual quantum memories. (Details are discussed in the Supplementary Information.) The ratio of the sensitivities with and without the virtual quantum memories is taken by $\sqrt{MT_{2p}/T_2}$, where the number of virtual quantum memories ($M$) composed of the dressed states and $T_{2p}$ is a $T_2$ of the dressed states. It should be noted that $2M$ corresponds to the number of dressed states.

Finally, we measured $T_{2p}$ and $T_2$ and estimated the $\sqrt{MT_{2p}/T_2}$. We measured those of a single NV center in a $^{12}$C enriched diamond because a coherent oscillation due to $^{13}$C nuclear spins on echo measurements $^{29}$ can be suppressed. The top of Fig. 6 shows the pulse sequence for the $T_{2p}$ and $T_2$ measurements. They were performed with or without a cw drive field. In these measurements, a phase cycle was applied to the last $\pi/2$ pulse indicated by $\pm$ signs in order to subtract artificial and dynamical contributions $^{30}$. In the case of a probe and drive power of $\sim 0.43$ MHz and $\sim 1.2$ MHz, respectively, the result of $T_{2p}$ (black plots) and $T_2$ (red plots) measurements with fitted decay curves are shown in Fig. 6. Consequently, we observed a coherence time of $T_{2p} \sim 1.5$ ms for the dressed states, which is more than two orders of magnitude longer than $T_2 \sim 4.2 \mu s$ of the undressed states. Such an extension can also be demonstrated by a dynamical decoupling technique, e.g., a Carr-Purcell-Meiboom-Gill (CPMG) sequence in the NV centres $^{6, 31-34}$. It should be noted that an extension of two orders of magnitude is much larger than the extension of about one order of $T_2$ in the dynamical decoupling techniques $^{6, 31-34}$. The proposed hybrid sensing technique may be suitable for an ensemble system, which has very short $T_2$.

In the case of an extension of more than two orders of magnitude, the sensitivity is approximately enhanced 27 times with the generation of a minimum number of dressed states ($M = 2$). In addition, the sensitivity can be effectively enhanced by increasing $M$ with a high-power driving field. Our model is, therefore, applicable to the realisation of the higher sensitivity on AC-field sensing.
CONCLUSION

In conclusion, we propose a hybrid quantum sensing with virtual quantum memories based on the dressed states. We experimentally demonstrate the generation of two dressed states of an NV centre in diamond by irradiating a drive field based on ATS and observe the generated dressed states with the theoretical prediction. We also estimate the enhancement of the sensitivity for AC magnetic field sensing. With the experimentally observed $T_{2p}$ and $T_2$, we show that the sensitivity can be enhanced at least one-order of magnitude. Our model should be applicable to realise hybrid quantum sensing using dressed states as virtual quantum memories, opening up a new sensing technique and inspiring quantum technologies.

Note: Recently, we have become aware of related works on quantum sensing with a one-time measurement based on a combination of the Mollow triplet and dynamical decoupling under sensing of a weak AC field with GHz frequencies $^{[35,36]}$. Our work has three differences: 1) the frequency range for the sensing target, 2) the quantum-memory effect for a higher sensitivity, and 3) robustness against environmental noises. Our work can realise sensing of a weak low-frequency AC field based on virtual quantum memories by dressed states generated by ATS.

METHODS

Sample Preparation

To generate dressed states by ATS, we used high-temperature and high-pressure (HTHP) type IIa (111) diamond. Nitrogen ($^{14}$N) was implanted into the diamond with a 30-kV accelerating energy. After implantation, the sample was annealed at 750 °C for 30 min. For the $T_2$ and $T_{2p}$ measurements, we used a single NV centre in a CVD-grown $^{12}$C enriched diamond layer on a type Ib (111) diamond substrate. The NV centres were generated during the growth of the diamond layer.

* Home-made confocal microscope with an electromagnetic field irradiation system

All experiments were performed by a homemade confocal microscope with an electromagnetic field (emf) irradiation system at room temperature. Figure 2(a) shows a schematic of the confocal-microscope setup with the emf irradiation system. A 532-nm laser focused by an objective lens illuminates an NV centre in diamond. The detection system is composed of a 50:50 beam splitter (BS) and two avalanche photodiodes (APDs) in order to detect the photoluminescence and measure $g^{(2)}(\tau)$. Two high-frequency oscillators, which are around 2.8 GHz, irradiate emfs to manipulate the electron spin of an NV centre. These correspond to the electron spin resonances of the NV centre on a static magnetic field generated by a neodymium magnet. The emfs are irradiated by a thin copper wire with a diameter of 10 μm.

AUTHORS CONTRIBUTION

HM, TT, and DM performed the measurements and the data analysis. HK synthesised the $^{12}$C-enriched diamond layers. All the authors contributed to the data analysis, discussion, and manuscript preparation.

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SUPPLEMENTARY INFORMATION

Estimation of the sensitivity of an NV magnetometer with virtual quantum states

We estimate the sensitivity of the weak AC magnetic field using our hybrid quantum sensing strategy. The key to our sensing is that the information of the AC magnetic field is accumulated in virtual quantum memories based on the dressed states. By using the fact that the dressed states have the same resonant frequencies, several dressed states generated for our sensing as virtual memories and quantum adder [S1] are then used to boost the sensitivity of the target.

Fig. S1(a) shows the sequence to demonstrate hybrid quantum sensing with virtual quantum states for the weak AC magnetic field. This sequence is considered as a combination of generation of the dressed states and conventional weak AC magnetic field sensing with a Hahn echo sequence, as described in Fig. S1(b) [S2]. The details of the sequence described in Fig. S1(a) are as follows: after initialisation of an NV electron spin by a pulsed laser, applications of a first $\pi/2$ pulse and pulsed strong mw-driving field generate dressed states in the NV centre. Under the generation of the dressed states, information about the AC magnetic field sensing is stored in these states by using the Hahn echo sequence. Finally, a quantum adder is used to integrate the phase in the stored information, as shown in Fig. S2.

The quantum adder consists of a Hadamard gate $H$ and a unitary operator $U^n$ of $|\Psi\rangle$. The $|\Psi\rangle$ works as an ancilla state, and the information in $n$ dressed states ($|\phi_n\rangle$) is transferred to $|\Psi\rangle$. For example, $|-1,0\rangle$ of an NV centre can be used for an ancilla state under the irradiation of drive field between $|-1,1\rangle$ and $|0,1\rangle$ of the NV centre described in Fig. S1(a) in the main text. In this way, this sensing can be done.

The minimum detectable value of the magnetic sensor ($B_{\text{min}}$) is given by the following relation: $B_{\text{min}} \propto 1/\sqrt{NT^2}$, where $N$ and $T_2$ are the numbers of NV centre (number of qubits) and $T_2$ of the NV electron spin, respectively [S2]. In the case of magnetometry with a single NV centre, $N = 1$. In our experiments, we observed $T_{2\rho} \sim 1.5$ ms and $T_2 \sim 4.2 \mu$s with and without the generation of dressed states in Fig. 6 of the main paper, respectively. It should be noted that at least four dressed states are generated in the $T_{2\rho}$ measurement. For example, we can confirm that the sensitivity is enhanced approximately 27 times using these values, and this relation with $N = M$ when $2M$ virtual states can be prepared by an irradiation of a strong mw-driving field.

Fig. S2. Quantum circuit of quantum Adder.

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