Nonlinear light-matter interaction in diamond

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The measured cavity mode response exhibits strong nonlinearity near a spin resonance. Data is compared with theoretical predictions and a good agreement is obtained. The nonlinear effect under study in the current paper is expected to play a role in any cavity-based magnetic resonance imaging technique and to impose a fundamental limit upon its sensitivity.

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Cavity quantum electrodynamics (CQED) [1] is the study of the interaction between photons confined in a cavity and matter. CQED has applications in a variety of fields, including magnetic resonance imaging and quantum computation [2]. The CQED interaction can be probed by measuring the response of a cavity mode. Commonly, the effect of matter on the response diminishes as the energy stored in the cavity mode under study is increased. This nonlinear effect, which is the focus of the current study, imposes a severe limit upon the performance of a variety of CQED systems.

In the current study we explore nonlinear CQED interaction between defects in a diamond crystal and a superconducting microwave cavity (resonator) having a spiral shape [3, 4]. Two types of defects are investigated, a negatively charged nitrogen-vacancy NV− defect and a nitrogen 14 (nuclear spin 1) substitutional defect (P1). Strong coupling between defects in diamond and a superconducting resonator has been demonstrated at ultra-low temperatures [5–11], however the regime of nonlinear response was not addressed. In this study, we find that the cavity response becomes highly nonlinear near a CQED resonance. In addition, for the case of NV− defects, the response is strongly affected by applying optically-induced spin polarization (OISP). The experimental findings are compared with theory and good agreement is obtained.

The experimental setup is schematically depicted in Fig. 1(a). Defects in a [100] type Ib diamond are created using 2.8 MeV electron irradiation with a dose of approximately $8 \times 10^{18}$ e/cm$^2$, followed by annealing at 800°C for 8 hours and acid cleaning, resulting in the formation of NV− defects with density of $1.23 \times 10^{17}$ cm$^{-3}$ [12]. The diamond wafer is then placed on top of a sapphire wafer supporting a superconducting spiral resonator made of niobium [see Fig. 1(b)]. Externally applied magnetic field $\textbf{B}$ is employed for tuning the system into a CQED resonance. A coaxial cable terminated by a loop antenna (LA) transmits both injected and off-reflected microwave signals. The LA has a coupling given by $\gamma_f/2\pi = 0.367$ MHz to the spiral's fundamental mode, which has a frequency of $\omega_c/2\pi = 2.53$ GHz and an unloaded damping rate of $\gamma_c/2\pi = 0.253$ MHz. All measurements are performed at a base temperature of $T = 3.1$ K. A network analyzer (NA) measurement of the temperature dependence of the resonance lineshape is seen in Fig. 1(c). The color-coded plot depicts the reflectivity coefficient $R_c = P_r/P_p$ in dB units, where $P_p = -70$ dBm ($P_r$) is the power of the injected (off-reflected) signal, as a function of both frequency of injected signal $\omega_\text{in}/2\pi$ and temperature $T$. Laser light of wavelength $\lambda_L = 532$ nm and intensity $I_L$ (in units of power per unit area) is injected into the diamond wafer using a multimode optical fiber F1, and another multimode optical fiber F2 delivers the emitted photoluminescence (PL) to an optical spec-

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FIG. 1: The experimental setup. (a) A loop antenna (LA) is coupled to the spiral resonator. Two multimode optical fibers are coupled to the diamond wafer. Fiber F1 is employed for delivering laser light of wavelength $\lambda_L = 532$ nm, and fiber F2 probes the emitted photoluminescence (PL). A magnetic field $B$ is applied parallel to the wafers surface using a superconducting coil magnet. (b) SEM image of the spiral resonator with 3 turns, an inner radius of 0.59 mm, an outer radius of 0.79 mm and thickness of 200 nm. (c) The resonance lineshape of the spiral's fundamental mode vs. temperature. (d) The magnetic induction magnitude $|\textbf{B}(r)|$ of the fundamental mode vs. position $r$ in a plane perpendicular to both wafers that contains the center of the spiral.
trum analyzer [see Fig. 2(a)]. Numerical calculation is employed for evaluating the shape of the spiral’s fundamental mode [see Fig. 1(d)].

The negatively-charged NV\textsuperscript{−} defect in diamond consists of a substitutional nitrogen atom (N) combined with a neighbor vacancy (V)\textsuperscript{13}. The ground state of the NV\textsuperscript{−} defect is a spin triplet having symmetry \(3 A_2\)\textsuperscript{14, 15}, composed of a singlet state \(|m_e = 0\rangle\) and a doublet \(|m_e = \pm 1\rangle\). The angular resonance frequencies \(\omega_{\pm}\) corresponding to the transitions between the state \(|m_e = 0\rangle\) and the states \(|m_e = \pm 1\rangle\) are approximately given by\textsuperscript{16, 18}

\[
\omega_{\pm} = D \pm \sqrt{\gamma_e^2 B_{||}^2 + E^2} + \frac{3 \gamma_e^2 B_{\perp}^2}{2D} , \quad (1)
\]

where \(B_{||}\) is the magnetic field component parallel to the axis of the NV defect and \(B_{\perp}\) is the transverse one. The parameter \(\gamma_e = 2\pi \times 28.03 \text{GHz} T^{-1}\) is the electron spin gyromagnetic ratio. In the absence of strain and when the externally applied magnetic field vanishes one has \(\omega_{\pm} = D\), where \(D = 2\pi \times 2.87 \text{GHz}\). Internal strain, however, may lift the degeneracy between the states \(|m_e = -1\rangle\) and \(|m_e = +1\rangle\), and give rise to a splitting by \(2E\). In a single crystal diamond the NV defects have four different possible orientations with four corresponding pairs of angular resonance frequencies \(\omega_{\pm}\).

The technique of optical detection of magnetic resonance (ODMR) can be employed for measuring the resonance frequencies \(\omega_{\pm}\)\textsuperscript{12, 20}. The measured PL spectrum is seen in Fig. 2(a). The integrated PL signal in the band 660 nm–760 nm is plotted as a function of microwave input frequency \(\omega_p/2\pi\) and externally applied magnetic field \(|B|\) in Figs. 2(b)-(c). In this measurement the microwave input power is set to \(P_p = 20 \text{dBm}\). The direction of the externally applied magnetic field \(B\) is found by fitting the measured ODMR frequencies \(\omega_{\pm}\) with the calculated values given by Eq. (1).

The ODMR spectrum contains a profound resonance feature at the frequency of the spiral resonator \(\omega_s/2\pi = 2.53 \text{GHz}\) [see Fig. 2(b)]. This feature is attributed to heating-induced change in the internal stress in the diamond wafer. Two (out of four) resonance frequencies \(\omega_s/2\pi\) can be tuned close to the spiral resonator frequency \(\omega_s/2\pi\) by setting the magnetic field \(|B|\) close to the value of 16 mT. In this region, which is magnified in Fig. 2(c), the ODMR becomes significantly deeper.

The same two spin resonances seen in Fig. 2(c) can be detected without employing the technique of ODMR provided that their frequencies are tuned close to the spiral resonator frequency \(\omega_s/2\pi\). The plots (D: P1; L0), (D: P2; L0) and (D: P3; L0) of Fig. 3 depict the measured values of the microwave reflectivity coefficient \(R_c\) with three different values of the injected signal microwave power \(P_p\). No laser light is injected into the diamond wafer in these measurements (labeled by L0 in Fig. 3). Henceforth this method of spin detection is referred to as cavity-based detection of magnetic resonance (CDMR). Both CDMRs seen in Fig. 3 exhibit strong dependence on \(P_p\), indicating thus that the interaction with the spins makes the cavity response highly nonlinear.

To account for the observed spin-induced nonlinearity, the experimental results are compared with theoretical predictions\textsuperscript{21}. The decoupled cavity mode is characterized by an angular resonance frequency \(\omega_c\), Kerr coefficient \(K_c\), linear damping rate \(\gamma_c\) and cubic damping (two-photon absorption) rate \(G_c\). The response of the decoupled cavity in the weak nonlinear regime (in which, nonlinearity is taken into account to lowest non-vanishing order) can be described by introducing the complex and mode amplitude dependent cavity angular resonance frequency \(\gamma_c\), which is given by

\[
\gamma_c = \omega_c - i\gamma_c + (K_c - iG_c) E_c , \quad (2)
\]

where \(E_c\) is the averaged number of photons occupying the cavity mode. The imaginary part of \(\gamma_c\) represents the effect of damping and the terms proportional to \(E_c\) represent the nonlinear contribution to the response.

The effect of the spins on the cavity response in the weak nonlinear regime is theoretically evaluated in\textsuperscript{22}. The steady state cavity mode response is found to be equivalent to the response of a mode having effective complex cavity angular resonance frequency \(\gamma_{\text{eff}}\) given by

\[
\gamma_{\text{eff}} = \gamma_c + \gamma_s , \quad (3)
\]

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and the cavity mode, \( T_{1,n} \) and \( T_{2,n} \) are the spin’s longitudinal and transverse relaxation times, respectively, \( \Delta_n = \omega_n - \omega_{n,n} \) is the frequency detuning between the cavity frequency \( \omega_n \) and the spin’s transition frequency \( \omega_{n,n} \), and \( P_{s,n} \) is the spin’s longitudinal polarization. The term proportional to \( E_c \) in the denominator of Eq. (3) gives rise to nonlinear response.

The coupling coefficients \( g_n \) can be extracted from the numerically calculated magnetic field induction \( B_c (r) \) of the spiral’s fundamental mode [see Fig. 1(d)] using the expression \( g_n = \gamma_c [B_c (r_n)] \sin \varphi_n / E_c^{1/2} \), where \( B_c (r_n) \) is the cavity mode magnetic induction at the location of the spin \( r_n \) and \( \varphi_n \) is the angle between \( B_c (r_n) \) and the NV axis. When all contributing spins share the same detuning factor \( \Delta \), polarization \( P_{s,n} \) and the same relaxation times \( T_1 \) and \( T_2 \), and when the variance in the distribution of \( g_n \) is taken into account to lowest nonvanishing order only, one finds that

\[
\Upsilon_s = \frac{N_{eff} g^2}{\Delta} \left( 1 + \frac{1}{\Delta T_s} \right),
\]

where \( \rho_s (r) \) is the density of contributing NV\(^-\) defects, \( N_{eff} = \int dr \rho_s P_{s,n} \) is their effective number and the effective coupling coefficient \( g_s \) is given by

\[
g^2_s = \frac{\gamma_c^2 \mu_0 \hbar \omega_c}{8 \pi I} \int dr |B_c|^2 \sin^2 \varphi P_{s,n} \int dr |B_c|^2 \int dr \rho_s P_{s,n}.
\]

In general, the averaged number of photons \( E_c \) is found from the steady state solution of the equations of motion that govern the dynamics of the system [22]. To lowest non-vanishing order in the coupling coefficient \( g_s \) the effect of spins can be disregarded in the calculation of \( E_c \). When, in addition, the intrinsic cavity mode nonlinearity, which is characterized by the parameters \( K_c \) and \( G_c \), has a negligibly small effect, the number \( E_c \) can be approximated by the following expression [see Eq. (37) in [26]]

\[
E_c = \frac{4^2 \mu_0 \hbar \omega_c (\omega_p - \omega_c)^2 + (\gamma_t + \gamma_c)^2}{8 \pi I}.
\]

As can be seen from Eq. (4), \( |\Upsilon_s| \) is a monotonically decreasing function of \( E_c \). This suggests that the approximation in which Eq. (6) is employed for evaluating \( E_c \) (without taking into account both nonlinearity and the coupling to the spins) remains valid even when \( 4g^2_s T_1 T_2 E_c \gg 1 \) provided that intrinsic cavity mode nonlinearity remains sufficiently small. When intrinsic cavity mode nonlinearity can be disregarded the cavity mode reflectivity \( R_c \) is given by

\[
R_c = \frac{(\omega_p - \Omega_c)^2 + (\gamma_t - \Gamma_c)^2}{(\omega_p - \Omega_c)^2 + (\gamma_t + \Gamma_c)^2},
\]

where the real frequencies \( \Omega_c \) and \( \Gamma_c \) are related to the complex frequency \( \Upsilon_{eff} \) by the relation \( \Upsilon_{eff} = \Omega_c - i \Gamma_c \).

![FIG. 3: Cavity mode reflectivity \( R_c \) with NV\(^-\) defects for various values of injected microwave power \( P_w \) (P1 = -90 dBm, P2 = -70 dBm and P3 = -60 dBm) and laser intensity \( I_L \) (L0 = 0, L1 = 5.6 mW mm\(^{-2}\), L2 = 12.8 mW mm\(^{-2}\) and L3 = 30 mW mm\(^{-2}\)). For each pair the top plot is experimental data (labeled by D) and the bottom is the theoretical prediction (labeled by F). The parameters used for the calculation for the case of laser on (off) are: \( P_{ST} = -0.035 \), \( P_{SO} = -0.55 \), \( \rho_s = 1.23 \times 10^{17} \) cm\(^{-3}\) [12], \( T_1 = 219 \) ns, \( T_{1T} = 23 \) ms \( (T_{1T} = 565 \) ms [23, 24]) and \( \omega_c / 2\pi = 0.43 \) Hz. The rate \( T_{1O}^{-1} = 0.16 \times \gamma_0 \), where \( \gamma_0 = I_L \sigma \omega_c / hc \) is the rate of optical absorption, where \( \sigma = 3 \times 10^{-17} \) cm\(^2\) [22] is the optical cross section, \( h \) is the Plank’s constant and \( c \) is the speed of light in vacuum. The effective coupling coefficient \( g_s \) for both cases of laser on and off is calculated using Eq. (5) and the numerically calculated mode shape [see Fig. 1(d)]. The volume inside the diamond wafer illuminated by the laser is 0.76 mm\(^3\).](image-url)
conserved in the optical dipole transitions between the triplet ground state \( ^3A_2 \) of NV− and the triplet first excited state \( ^3E \). However, transition from the spin states \( m_e = \pm 1 \) of \( ^3E \) to the ground state is also possible through an intermediate singlet states in a two-steps non-radiative process. Such non-radiative process is also possible for the decay of the state \( m_e = 0 \) of \( ^3E \), however, the probability of this process is about 7 times smaller than the probability of non-radiative decay of the \( m_e = \pm 1 \) states \[13\]. The asymmetry between the decay of \( m_e = 0 \) state, which is almost exclusively radiative, and the decay of the states \( m_e = \pm 1 \), which can occur via non-radiative process, gives rise to OISP. For our experimental conditions the probability to find any given NV− defect at any given time not in the triplet ground state \( ^3A_2 \) is about \( 10^{-5} \) or less \[13\]. This fact is exploited below for taking the effect of OISP into account within the framework of a two-level model.

The effect of OISP can be accounted for by adjusting the values of the longitudinal relaxation time \( T_z \) and longitudinal steady state polarization \( P_z \) and make them dependent on laser intensity \( I_L \). The total rate of spin longitudinal damping \( \gamma_1 \) is given by

\[
\gamma_1 = -\frac{P_z - P_{z\text{ST}}}{T_{1T}} - \frac{P_z - P_{z\text{SO}}}{T_{1O}}, \tag{8}
\]

where the first term represents the contribution of thermal relaxation and the second one represents the contribution of OISP. Here \( P_z \) is the instantaneous longitudinal polarization and \( T_{1T}^{-1} (T_{1O}^{-1}) \) is the rate of thermal relaxation (OISP). In steady state and when \( T_{1T}^{-1} > T_{1O}^{-1} \) (i.e. when OISP is negligibly small) the coefficient \( P_{z\text{ST}} = -\tanh(\omega_p/2k_B T) \) is the value of \( P_z \) in thermal equilibrium, where \( k_B \) is the Boltzmann’s constant and where \( T \) is the temperature. In the opposite limit of \( T_{1O}^{-1} > T_{1T}^{-1} \) (i.e. when thermal relaxation is negligibly small) the coefficient \( P_{z\text{SO}} \) is the value of \( P_z \) in steady state. Note that the total longitudinal damping rate \( \gamma_1 \) can be expressed as \( \gamma_1 = -T_{1T}^{-1}(P_z - P_{z\text{ST}}) \), where \( T_{1T}^{-1} = T_{1T}^{-1} + T_{1O}^{-1} \) is the effective longitudinal relaxation rate, and the effective steady state longitudinal polarization \( P_{z\text{ST}} \) is given by \( T_{1T}^{-1}P_{z\text{ST}} = T_{1T}^{-1}P_{z\text{ST}} + T_{1O}^{-1}P_{z\text{SO}} \).

The theoretical expressions given above for \( T_{1T}^{-1} \) and \( P_{z\text{ST}} \) are employed for generating the plots labeled by F of Fig. 3 for both cases of laser off (L0) and laser on (L1, L2 and L3). In spite of the simplicity of the model that is employed for the description of OISP, good agreement is obtained from the comparison with the CDMR data plots labeled by D in a very wide range of values for the microwave power and laser intensity (the entire explored range of \( P_p < 0 \) dBm and \( I_L < 30 \) mW mm\(^{-2} \)).

A CQED resonance due to P1 defects \[32, 33\] is observed when the externally applied magnetic field is tuned close to the value of 89 mT (see Fig. 4). When both nuclear Zeeman shift and nuclear quadrupole coupling are disregarded, the spin Hamiltonian of a P1 defect is given by \[32, 34, 35\]

\[
\hat{H} = \gamma_e \mathbf{B} \cdot \mathbf{S} + \hbar^2 A_{1}\left(S_x I_x + S_y I_y\right) + \hbar^2 A_{10} S_z I_z, \quad \mathbf{S} = (S_x, S_y, S_z)
\]

is an electronic spin 1/2 vector operator, \( \mathbf{I} = (I_x, I_y, I_z) \) is a nuclear spin 1 vector operator, \( A_\parallel = 2\pi \times 114.03 \text{MHz} \) and \( A_\perp = 2\pi \times 81.33 \text{MHz} \) are respectively the longitudinal and transverse hyperfine parameters, and the \( z \) direction corresponds to the diamond (111) axis. When the externally applied magnetic field \( \mathbf{B} \) is pointing close to a crystal direction (100), i.e. when \( \cos^2 \theta \approx 1/3 \), the electron spin resonance at angular frequency \( \gamma_e B \) is split due to the interaction with the nuclear spin into three resonances, corresponding to three transitions, in which the nuclear spin projection along the \( z \) axis is conserved. To first order in perturbation theory the resonance frequencies are given by \( \gamma_e B \pm \omega_{\text{en}} \), where \( \gamma_e B + \omega_{\text{en}} \) and \( \gamma_e B - \omega_{\text{en}} \), where \( \omega_{\text{en}}^2 = A^2 \cos^2 \theta + A^2_1 \sin^2 \theta \). For the case where \( \cos^2 \theta = 1/3 \) the calculated splitting is given by \( \omega_{\text{en}}/2\pi = 93.5 \text{MHz} \), whereas the value extracted from the data seen in Fig. 4 is 93.82 MHz. The plots labeled by F in Fig. 4 represent the theoretical prediction based on the analytical expressions \[41\], \[42\] and \[77\]. The comparison with the CDMR data plots (labeled by D) yields a good agreement. The parameters that have been employed for the calculation are listed in the figure caption.

The nonlinearity in cavity response has an important impact on sensitivity of spin detection. Let \( S_N \) be the minimum detectable change in the number of spins \( \delta N \) per a given square root of the available bandwidth (i.e. the inverse of the averaging time). When the cavity’s response is linear \( S_N \) is proportional to \( E_{\text{se}}^{-1/2} \) [see Eq. (1) in \[37\]] and thus in this regime sensitivity can be enhanced by increasing the energy stored in the cavity \( E_{\text{se}} \). However, nonlinearity, which can be avoided only when \( E_c \ll E_{\text{se}} \equiv (4g_e^2 T_2^1)^{-1} \) [see Eq. (1)], imposes a bound upon sensitivity enhancement. When the sensitivity coefficient \( S_N \) is calculated according to Eq. (1) in Ref. \[37\] for the case where the number of cavity photons
is taken to be $E_{cc}$, one finds that $S_N$ becomes

$$S_N \approx \frac{2}{|P_{ST}|^{3/2}} \left( \frac{\gamma S T_1}{g_2^2 T_2} \right)^{1/2}.$$  \tag{9}

Note that in general $2T_1/T_2 \geq 1$ [see Eq. (A79) in [22]]. For example, for the parameters of our device with laser off Eq. (9) yields $S_N = 5 \times 10^7 \text{Hz}^{-1/2}$. The estimate given by Eq. (9) is expected to be applicable for any cavity-based technique of spin detection.

To conclude, in this work we have observed strong coupling between a superconducting microwave cavity and spin ensembles in diamond (the measured values of the cooperativity parameter $N_{eff} g_2^2 / \gamma g_2$ are 14 with the NV$^-$ ensemble and laser intensity of 30 mW mm$^{-2}$ and 6.2 with the P1 ensemble). We find that the coupling imposes an upper bound upon the input microwave power, for which the cavity response remains linear. This bound has important implications on the sensitivity of traditional spin detection protocols that are based on linear response. On the other hand, in some cases nonlinearity can be exploited for sensitivity enhancement (e.g. by generating parametric amplification). However, further study is needed to explore ways of optimizing the performance of sensors operating in the nonlinear regime.

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