Coupling of diamond nanocrystals to a high-$Q$ whispering-gallery microresonator

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We theoretically investigate a cavity quantum electrodynamics system in which a high-$Q$ whispering-gallery-mode (WGM) microresonator interacts with multiple diamond nanocrystals each containing a single nitrogen-vacancy (NV) center. With a tapered fiber waveguide coupling the WGMs, the transmission spectrum of the system exhibits a twofold-splitting phenomenon, where the first fold of splitting originates from the nanocrystals’ Rayleigh scattering and the second fold stems from the NV center dipole coupling with the WGMs. The interacting system can serve as a platform to generate quantum entanglement between two NV centers embedded in distant diamond nanocrystals. Our further study reveals that the Rayleigh scattering of the nanocrystals plays a positive role in generating entanglement in some cases. The analytical results show that the relatively strong scattering leads to a larger concurrence for the fixed dipole-cavity detunings. This investigation may hold potential for solid-state quantum information processing applications.

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1. INTRODUCTION

Strong light-matter interaction within the context of cavity quantum electrodynamics (CQED) is of central concern in various fundamental studies and applications [1–4]. Among CQED platforms, combining high-$Q$ whispering-gallery-mode (WGM) microcavities and nitrogen-vacancy (NV) centers in diamond is quite promising. Therein, WGM microcavities possess ultrahigh $Q$ factors and small mode volumes, enabling strong temporal and spatial confinement of photons [5–12]. The massive on-chip fabrication and excellent scalability also make them promising for real applications. Meanwhile, NV centers in diamond can be used as a long-lived memory with optical readout, which is especially desirable in quantum information processing (QIP) [13–20]. Therefore, coupling NV centers to WGM microcavities provides an excellent platform for CQED studies and QIP applications [21–24]. Experimentally, the efficient coupling of NV center to a microcavity has been demonstrated [25–30].

One of the features of NV-WGM CQED systems is that the quantum emitters (NV centers) are embedded inside the diamond nanocrystals, whose sizes are much larger than the neutral atoms and ions used in conventional CQED systems. Thus, when diamond nanocrystals are coupled to ultrahigh-$Q$ microcavities, their scattering of light should be taken into consideration. In our previous work, we revealed that the Rayleigh scattering of a nanocrystal plays an important role in photon transport properties by exploring the interaction between a WGM microcavity and a diamond nanocrystal containing a single NV center [31]. In this paper, we investigate the multiparticle solid-state CQED system, which consists of a WGM microcavity and multiple diamond nanocrystals each containing a single NV center. We reveal that the system generally has four eigenmodes and the eigenfrequencies exhibit a twofold-splitting phenomenon. This phenomenon is a result of the combination of the multiple-Rayleigh-scatterer-induced mode splitting [32–35] and the dipole-induced mode splitting [31,36]. We also find that the Rayleigh scattering of the nanocrystals plays a positive role in generating entanglement between two NV centers surrounding the microcavity.

The paper is organized as follows. In Sec. II, we establish the quantum model for the coupling system. In Sec. III, we explore the steady-state field and the eigenmodes of the system. Section IV presents the transmission for the two identical nanocrystals case. In Sec. V, we study the entanglement of two NV centers via simultaneous coupling to the WGMs. Finally, a summary is presented.

II. MODEL

Figure 1 shows the coupling system where $n$ diamond nanocrystals each containing a single NV center are placed on the surface of a toroidal microcavity [37]. The microcavity supports a pair of WGMs with counterpropagating directions, clockwise (cw) and counterclockwise (ccw). A tapered fiber is used to excite the cavity modes and collect their emissions [38]. The Hamiltonian can be divided into two parts

$$H = H_s + H_r,$$

where $H_s$ is the system Hamiltonian and $H_r$ is the Hamiltonian related to the reservoir. The system Hamiltonian $H_s$ includes two types of interactions: the dipole coupling between the WGMs and the NV centers, and the coupling between the WGMs induced by the Rayleigh scattering of the nanocrystals. Under the rotating wave approximation, the system Hamiltonian $H_s$ can be written as (in the units of $\hbar = 1$)

$$H_s = \omega_c \sum_{l=1}^{n} |e_l \rangle \langle e_l | + \omega_c \sum_{m} \hat{a}_m^\dagger \hat{a}_m + \sum_{l=1}^{n} \sum_{m=1}^{\infty} (G_{l,m} \hat{a}_m^\dagger e^{i \omega_c t} \hat{a}_{l-} + \text{H.c.}) + \sum_{l=1}^{n} \sum_{m,n} g_{l,m,n} \hat{a}_m^\dagger e^{i \omega_c t} \hat{a}_n e^{-i \omega_c t}. \quad (2)$$
Here, the first two terms describe the free Hamiltonian of the NV centers and the WGMs, with the resonant frequency $\omega_c$ and $\omega_s$, respectively; $\sigma_l = |\psi_l\rangle \langle \psi_l|$ stands for the descending operator of the $l$th NV center zero-phonon line transition (637 nm); $a_m \ (a_m^\dagger)$ denote annihilation (creation) operators of the $m$th cavity modes ($m = cw, ccw$). $\xi_{l,m}$ represents the phase of the $m$th cavity mode on the position of the $l$th NV center, with $\xi_{l,cw} = -\xi_{l,ccw}^\ast = \xi_l$. The third term characterizes the dipole interactions between the NV centers and the WGMs, with the coupling strength $G_{l,m} \equiv G_l = \mu \sqrt{\omega_c/(2\hbar\epsilon_l\varepsilon_{\text{eff}}V_c)} f_c(\tilde{r}_l)$. The last term characterizes the nanocrystal-induced scattering among the nanocrystals and the WGMs, with the coupling strengths $g_{l,m,m'} \equiv -g_l = -\alpha_f f_c^2(\tilde{r}_l)\omega_c/(2V_c)$ [39]. Here $f_c(\tilde{r}_l) = |E(\tilde{r}_l)/E_{\text{max}}\rangle$ is the normalized field distribution function of the $l$th NV center at the position $\tilde{r}_l$; $V_c$ is mode volumes of the cavity modes; $\mu = 2.74 \times 10^{-20}$ C · m is the dipole moment of the NV center zero-phonon line transition; $\alpha_f = 4\pi R_l^3 (\epsilon_d - \epsilon_s)/ (\epsilon_d + 2\epsilon_s)$ represents the polarizability of the $l$th nanocrystal (spherical) with radius $R_l$; $\epsilon_d = 2.4^2$ ($\epsilon_s = 1^2$) denotes the relative permittivity of the surrounding medium; $\varepsilon_{\text{eff}}$ is the effective permittivity taking into account of the local-field effect [40]. $\varepsilon_{\text{eff}}$ is a function of the nanocrystals’ size $R_l$, and $\varepsilon_s < \varepsilon_{\text{eff}} < \varepsilon_d$. This local-field effect does not affect the scattering coefficients. To focus on the physics, we use $\varepsilon_{\text{eff}} = 1$ in our calculation.

The Hamiltonian related to reservoir is given by

$$H_r = \sum_{l=1}^n \sum_{j=1}^2 \sum_{k=1,2} \alpha_{l,j,k} b_{l,j,k}^\dagger b_{l,j,k}$$

$$+ \sum_{l=1}^n \sum_{j} (G_l b_{l,j}^\dagger \sigma_l + \text{H.c.})$$

$$+ \sum_{l=1}^n \sum_{m} (g_{l,m} a_m^\dagger e^{i\omega_m t} a_{l,1,2} + \text{H.c.}).$$

Here, the first term describes the free Hamiltonian of the NV centers, where $b_{l,j,k}^\dagger, b_{l,j,k}$ and $\omega_{l,j,k}$ denote the creation operator, annihilation operator and the resonant frequency of the $j$th reservoir modes, respectively. $k = 1, 2$ denote two kinds of reservoir, which interact with the NV centers and WGMs. The second term characterizes the dipole interactions between the NV centers and the reservoir modes with the coupling strengths $G_{l,j}$; the last term in $H_r$ characterizes the nanocrystal-induced scattering among the nanocrystals and the reservoir modes with the coupling strengths $g_{l,m,j}$. Note that we focus on the how the nanocrystal scattering affect the WGMs-NV center coupling, thus only these two types of interaction are important. Other interactions related to the diamond nanocrystal, such as coupling between the reservoir modes induced by the nanocrystal scattering, are negligible here since they do not directly affect the WGMs and NV centers.

Taking the input field and the cavity decays into account, the equations of motion for the system are given by [41]

$$\dot{a}_m = -i [a_m, H] - \frac{\kappa_0 + \kappa_1}{2} a_m - \sqrt{\kappa_0} a_m + \tilde{f}_m,$$

$$\dot{b}_{l,j,k} = -i [b_{l,j,k}, H], \quad \dot{\sigma}_l = -i [\sigma_l, H].$$

where $\kappa_0 \equiv \omega_c/Q_0$ is the intrinsic decay rate, $\kappa_1$ describes the fiber-WGM coupling strength, and $a_m^\ast$ denotes the input field. $\tilde{f}_m$ is the noise operator associated with $\kappa_0$. Using Weisskopf-Wigner approximation, the reservoir modes can be eliminated, yielding

$$\dot{a}_m = -i \tilde{\omega}_c a_m + i \sum_{l=1}^n \sum_{m'} \tilde{G}_{l,m} e^{i(\tilde{\omega}_m - \tilde{\omega}_c) t} a_{m'},$$

$$- i \sum_{l=1}^n \tilde{G}_{l} e^{i(\tilde{\omega}_c - \tilde{\omega}_s) t} \tilde{\sigma}_l - \sqrt{\kappa_0} a_m + \tilde{f}_m,$$

$$\dot{\sigma}_l = -i \tilde{\omega}_c \sigma_l + i G_l \sum_{m} e^{-i\omega_m t} a_m \sigma_{l,m} + \tilde{f}_l,$$

where the coupling of the NV centers and the reservoir results in the spontaneous decay of the excited state $|\psi_l\rangle$, with the rate $\gamma_l = \omega_c^2/3(\epsilon_d \epsilon_s \hbar^2 c) \sim 2\pi \times 13$ MHz, while the scattering of WGMs to the reservoir is modeled by the additional energy damping of the WGMs, with the rate $\kappa_{R,j} = \alpha_f^2 E_j^2 f_c^2(\tilde{r}_j)\omega_c^2/(6\pi c^3 V_c)$ [39]. Here $\sigma_l = |\psi_l\rangle \langle \psi_l| - |0\rangle \langle 0|$ is the Pauli operator of the $l$th NV center; $\tilde{f}_m$ and $\tilde{f}_l$ are the total noise operators for the WGM and the NV centers, respectively, whose precise forms do not matter and we can simply neglect them in the following discussion since at optical frequencies these operators act on the vacuum state; $\gamma_l = \omega_c/2 \kappa_0 (\kappa_0 + \kappa_1)/2$, $\omega_c = \omega_{\text{eff}} - i\gamma_l/2$ can be considered as the complex cavity resonant frequency and NV center resonant frequency, where the imaginary part of these complex frequencies represent the linewidths of the resonant modes; $\tilde{g}_l = g_l + i\kappa_{R,j}/2$ can also be viewed as the complex frequency shift induced by the nanocrystal scattering, in which the real part $g_l$ represents the scattering-induced frequency shift and the imaginary part $\kappa_{R,j}/2$ represents the scattering-induced linewidth broadening. Note that the rates $g_l$ and $\kappa_{R,j}$ are sensitive to the nanocrystal’s size $R_l$, where we have $g_l \propto R_l^2$ and $\kappa_{R,j} \propto R_l^3$. For small nanocrystals, the effect of their scattering can be neglected, while for large nanocrystals, the scattering can play an important in the system.
III. STEADY-STATE FIELD AND EIGENMODES

We assume that the input field $a_{in}$ is weak monochromatic field with frequency $\omega_p$, while $a_{in}$ is vacuum field. The weak-field excitation ensures that the average photon number inside the cavity is much less than one and the NV centers are predominantly in the ground state. By substituting $\sigma_z$ for its average value of $-1$, Eq. (7) can be linearized. In the steady state, we obtain the intracavity light field

$$a_{cw} = \frac{-i \left( \Delta_{pc} + \sum_{i=1}^{n} \eta_i \right) \sqrt{\kappa} a_{in}}{\left( \Delta_{pc} + \sum_{i=1}^{n} \eta_i \right)^2 - \Delta_{pc} + \sum_{i=1}^{n} \eta_i e^{2i\theta} \sum_{i=1}^{n} \eta_i e^{-2i\theta} + \hat{f}_{cw}},$$

$$a_{ccw} = \frac{i \left( \sum_{i=1}^{n} \eta_i e^{-2i\theta} \right) \sqrt{\kappa} a_{in}}{\left( \Delta_{pc} + \sum_{i=1}^{n} \eta_i \right)^2 - \Delta_{pc} + \sum_{i=1}^{n} \eta_i e^{2i\theta} \sum_{i=1}^{n} \eta_i e^{-2i\theta} + \hat{f}_{cw}},$$

where $\Delta_{pc} = \omega_p - \tilde{\omega}_c = \Delta_p + i \left( \kappa_0 + \kappa_1 \right) / 2$ is the complex detuning; $\eta_i = \tilde{g}_i - \tilde{G}_i$ is a parameter, which reveals that the effect of the diamond nanocrystal can be divided into two separate parts, in which $\tilde{g}_i$ represents nanocrystal-induced scattering and $\tilde{G}_i = G_i^2 / \Delta_{pc}$ represents NV center dipole coupling with $\Delta_{pc} = \omega_p - \tilde{\omega}_c$. $\hat{f}_{cw}$ and $\hat{f}_{ccw}$ are noise operators.

In the following we assume all the nanocrystals are identical with the same radius and field distribution function [i.e., $R_i \equiv R$, $f_i \equiv f_i$]. In this case $\tilde{g}_i \equiv \tilde{g}$, $\tilde{G}_i \equiv \tilde{G}$, $\eta_i \equiv \eta$, and the system can be described by four nontrivial eigenmodes, which are hybrid modes composed of $a_{cw}$, $a_{ccw}$, and $\sigma_z$, given by

$$a_{+}^{(1,2)} = \frac{1}{\sqrt{2}} \left( e^{i\theta} a_{cw} + e^{-i\theta} a_{ccw} \right) + \sqrt{2G} \sum_{i=1}^{n} \cos \left( \tilde{\omega}_0 - \tilde{\omega}_i \right) \sigma_z + \hat{f}_{+}^{(1,2)},$$

$$a_{-}^{(3,4)} = \frac{1}{\sqrt{2}} \left( e^{-i\theta} a_{cw} - e^{i\theta} a_{ccw} \right) - i \sqrt{2G} \sum_{i=1}^{n} \sin \left( \tilde{\omega}_0 - \tilde{\omega}_i \right) \sigma_z + \hat{f}_{-}^{(3,4)},$$

with the corresponding complex eigenfrequencies

$$\tilde{\omega}_+^{(1,2)} = \tilde{\omega}_c + \tilde{\omega}_e - \tilde{\omega}_0^{(1,2)} \pm \sqrt{(\tilde{\omega}_c - \tilde{\omega}_e - \tilde{\omega}_0^{(1,2)})^2 + 4\tilde{\omega}_0^{(1,2)} G^2},$$

$$\tilde{\omega}_-^{(3,4)} = \tilde{\omega}_c + \tilde{\omega}_e - \tilde{\omega}_0^{(3,4)} \pm \sqrt{(\tilde{\omega}_c - \tilde{\omega}_e - \tilde{\omega}_0^{(3,4)})^2 + 4\tilde{\omega}_0^{(3,4)} G^2},$$

$$\tilde{\omega}_+^{(1,2)} = \tilde{\omega}_c + \tilde{\omega}_e - \tilde{\omega}_0^{(1,2)} \pm \sqrt{(\tilde{\omega}_c - \tilde{\omega}_e - \tilde{\omega}_0^{(1,2)})^2 + 4\tilde{\omega}_0^{(1,2)} G^2},$$

$$\tilde{\omega}_-^{(3,4)} = \tilde{\omega}_c + \tilde{\omega}_e - \tilde{\omega}_0^{(3,4)} \pm \sqrt{(\tilde{\omega}_c - \tilde{\omega}_e - \tilde{\omega}_0^{(3,4)})^2 + 4\tilde{\omega}_0^{(3,4)} G^2},$$

where $\tilde{\omega}_0^{(1,2)} = \frac{1}{2} \arg \sum_{i=1}^{n} e^{2i\theta} \cdot \tilde{\omega}_0 = n \mp \tilde{\omega}_0 = \tilde{\omega}_0^{(1,2)}$. $\tilde{\omega}_0^{(3,4)}$ is the complex eigenfrequencies $\tilde{\omega}_0^{(p)}$ describe both the resonant frequency (real part) and the linewidth (imaginary part) of the $p$th eigenmode. We can find that $\tilde{\omega}_+^{(1,2)} < \tilde{\omega}_0^{(4)} < \tilde{\omega}_-^{(3)} < \tilde{\omega}_-^{(1,2)}$, where $\omega_+^{(p)} = \text{Re} \tilde{\omega}_0^{(p)}$ represents the resonant frequencies.

It is revealed from Eqs. (10)–(13) that “twofold-splitting” phenomenon occurs. The first fold of splitting originates from the coupling between the cw and ccw modes, forming two standing-wave modes $a_{\pm} = (e^{-i\theta} a_{cw} \pm e^{i\theta} a_{ccw}) / \sqrt{2}$, denoted by “+” mode and “−” mode. The second fold of splitting stems from the coupling between “±” modes and the NV centers, leading to polariton modes $a_{(p)}^{(p)} (p = 1, 2, 3, 4)$.

Note that these four eigenmodes in Eqs. (10) and (11) can be degenerated in some cases, depending on the relative position of the nanocrystals. Consider the following special situations:

(i) Totally constructive interference case ($\tilde{\eta} = n$). When $\tilde{\eta}_n = n$, the $n$ nanocrystals have the same standing wave phase. In this case $\tilde{\tilde{\omega}}_0^{(1,2)} = 2 n \tilde{\omega}_e$, which means the “+” mode and the “−” mode keep unchanged. Therefore, in the second fold of splitting, the “+” mode splits while the “−” mode keeps unchanged. There are three eigenmodes that can be excited, with the eigenfrequencies given by

$$\tilde{\omega}_+^{(1,2)} = \tilde{\omega}_c + \tilde{\omega}_e - 2 n \tilde{\omega}_e \pm \sqrt{(\tilde{\omega}_c - \tilde{\omega}_e - 2 n \tilde{\omega}_e)^2 + 4 n G^2},$$

and $\tilde{\omega}_-^{(3,4)} = \tilde{\omega}_+^{(1,2)}$.

In other general case, $0 < \tilde{\omega} < n$, no degeneracies exist and the system has four resonant eigenfrequencies.

IV. TRANSMISSION

To show the eigenmodes more explicitly, we calculate the transmission spectrum of the system with fiber taper coupling. Using the input-output formalism [41] $a_{cw}^{\text{out}} = a_{cw}^{\text{in}} + \sqrt{\kappa} a_{cw}$, we can calculate the transmitted field $a_{cw}^{\text{out}}$ and thereby the transmission $T = \left| a_{cw}^{\text{out}} / a_{cw}^{\text{in}} \right|^2$. We plot the transmission for various phase differences in Fig. 2, which verifies the three different regimes discussed in the
previous section. (i) For the phase difference \( \xi = q\pi \) \((q = 0, \pm 1, \pm 2, \ldots)\), we obtain \( \zeta = 2 \). There exist three resonant dips: the central dip representing the “−” mode decoupled with the NV center and the two side dips which are split due to the coupling between the “+” mode and the NV centers. (ii) For \( \xi = \pi/2 + q\pi, \zeta = 0 \), indicating that one nanocrystal is located at the antinode of a standing-wave mode, while the other nanocrystal is located at its node. In this case one nanocrystal interacts with the “+” mode while the other nanocrystal interacts with the “−” mode. The “±” modes are degenerate and the same splitting occurs. (iii) For \( \xi \neq q\pi/2, 0 < \xi < 2 \), the NV centers couple to both “±” modes, leading to a twofold splitting, and four resonant dips appear.

To further shed light on the physical properties, in Fig. 3 we present contour plots of the transmission for different nanocrystal sizes and phase differences. Figures 3(a)–3(c) show explicitly the effect of the nanocrystal-induced scattering. In all the three regimes, as the nanocrystals’ size increase, the modes shift to the red side band and the linewidths are broadened. For the totally constructive interference case \( \xi = q\pi \), especially, the “−” mode decouples with the NV centers, which keeps unchanged as the nanocrystals’ size increase, as illustrated in Fig. 3(a). The effect of the relative position of the nanocrystals is depicted in Fig. 3(d), where the nanocrystals’ size is fixed. We can see the “crossing” phenomenon occurs for \( \xi = q\pi \) and \( \xi = \pi/2 + q\pi \). Note that the outer two lines correspond to the “+” mode and the inner two correspond to the “−” mode. The “+” mode splits in all cases, while the “−” mode does not split for \( \xi = q\pi \). When \( \xi = \pi/2 + q\pi \), the “±” modes are degenerate and they have the same splitting.

\[ H_1 = \sum_{l=1}^{n} \left( \omega_{el} |e\rangle \langle e| + \omega_{l} |l\rangle \langle l| \right) + \sum_{m} \omega_{l} a_{m}^\dagger a_{m} \]

\[ + \sum_{l=1}^{n} \sum_{m} (g_{l} a_{m}^\dagger e^{i\phi_{m}} |0\rangle \langle e| + \text{H.c.}) \]

\[ + \sum_{l=1}^{n} \sum_{m} \Omega_{l} e^{i\omega_{l}t} |l\rangle \langle e| + \text{H.c.}) \]

\[ - \sum_{l=1}^{n} \sum_{m,m'} g_{l} a_{m}^\dagger e^{i\phi_{m}} a_{m'} e^{-i\phi_{m'}} \]  

(V. Entanglement)

After revealing the basic physics of this coupling system, we proceed to study its application in quantum information processing [42]. One major task in quantum computing and quantum cryptography is to generate entanglement [43]. Remarkably, we find that in our system the nanocrystal-induced scattering plays an important role in the entanglement evolution between NV centers. In Ref. [44], Jin et al. show that the rough surface of the microcavity can enhance the atomic entanglement by studying the entanglement of two atoms surrounding the microcavity. Our investigations reveal that the entanglement enhancement can be achieved by harnessing the scattering of the solid-state quantum system itself, which holds great potential for solid-state quantum information processing applications.

We consider the coupling between the microcavity and \( n \) identical nanocrystals each containing a single NV center. The qubits can be constructed from the ground states \( |0\rangle \) and \( |1\rangle \), which denotes the states \( |3/2 A, m_e = 0\rangle \) and \( |3/2 A, m_e = \pm 1\rangle \), respectively. The state \( |3/2 E, m_e = 0\rangle \) is labeled by \( |e\rangle \). The cavity modes have a detuning \( \Delta_{n} \) from the transition \( |0\rangle - |e\rangle \), and a strong drive field is applied to all the nanocrystals, which couples \( |1\rangle \) state and \( |e\rangle \) state.

The Hamiltonian of the system is given by
where $\omega_{1}/2\pi = 2.88$ GHz is the zero-field energy spacing between states $|1\rangle$ and $|0\rangle$, $\omega_L$ is the drive laser frequency and $\Omega_L$ is the Rabi frequency. Note that we no input field is used to excite the WGMs here. In the large detuning condition (i.e., $\Delta_{cc} = \omega_r - \omega_{1} - \omega_{L}$ are much larger than the coupling strength) we can adiabatically eliminate the excited state $|e_{i}\rangle$ and construct an effective two-level system, with the energy diagram shown in Fig. 4. According to the standard procedure of adiabatic elimination [41,45], we obtain the effective Hamiltonian

$$H_{\text{eff}} = \sum_{l=1}^{n} |1\rangle \langle 1| + \sum_{l=1}^{m} (G_{l}^* a_{l}^\dagger e^{i\xi_l n} |0\rangle \langle 1| + \text{H.c.}) - \sum_{l=1}^{m} g_{l} a_{l}^\dagger a_{l} e^{-i\xi_{l} n},$$

(19)

where the effective detuning $\Delta' = \Delta_{cc} - \Delta_{L} - \Omega_{L}^{2}/\Delta_{cc}$ (here we have dropped the negligible bilinear terms), the effective dipole coupling strength $G' = -\Omega_{L} G/\Delta_{cc}$ and the effective nanocrystal scattering strength $g' = g + G^{2}/\Delta_{cc}$. The master equation for the effective two-level system is given by

$$\dot{\rho} = -i[H_{\text{eff}}^\dagger, \rho] + \frac{K}{2} \sum_{l=1}^{n} [\mathcal{L}(a_{l}^\dagger e^{i\xi_l n}) + a_{l} e^{i\xi_{l} n} \rho + \mathcal{L}(a_{l}^\dagger) \rho + \mathcal{L}(a_{l}) \rho],$$

(20)

where $\mathcal{L}$ is the Lindblad form, with $\sigma_{r} = \{a_{l}^\dagger\}$ and $\gamma' = y_{r} \Omega_{L} G/\Delta_{cc}$ is the effective spontaneous decay rate between states $|1\rangle$ and $|0\rangle$.

First, we assume only one NV center (the $k$th) is in |1⟩ state and the others are in |0⟩ states, while the WGMs are in the vacuum state. It is found that the system can be divided into two subsystems (i.e., the ground-state subsystem and the excited-state subsystem). Thus the density operator can be expressed as $\rho = \rho_{00} |0\rangle \langle 0| \otimes \rho_{\text{ccw}} |0\rangle \langle 0| + |\Psi_{e}\rangle \langle \Psi_{e}|$, where

$$|\Psi_{e}\rangle = G e^{i\xi_{e}} \left[ \begin{array}{c} \cos(\xi_{0} - \xi_{e}) e^{-i\omega_{1,1}^{(3)}} - e^{-i\omega_{1,1}^{(4)}} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right] |1_{\text{cw}}\rangle |0\rangle^{\otimes(n+1)}$$

$$+ G e^{-i\xi_{e}} \left[ \begin{array}{c} \cos(\xi_{0} - \xi_{e}) e^{-i\omega_{1,1}^{(2)}} - e^{-i\omega_{1,1}^{(3)}} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right] |1_{\text{ccw}}\rangle |0\rangle^{\otimes(n+1)}$$

$$+ \sum_{l=1}^{n} \left[ \begin{array}{c} 2 \cos(\xi_{0} - \xi_{e}) \cos(\xi_{0} - \xi_{l}) \left( \begin{array}{c} \omega_{1,1}^{(3)} - \omega_{1,1}^{(4)} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right) e^{-i\omega_{1,1}^{(3)}} - e^{-i\omega_{1,1}^{(4)}} \right] |1_{l}\rangle |0\rangle^{\otimes(n+1)}$$

$$+ 2 \sin(\xi_{0} - \xi_{e}) \sin(\xi_{0} - \xi_{l}) \left( \begin{array}{c} \omega_{1,1}^{(3)} - \omega_{1,1}^{(4)} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right) e^{-i\omega_{1,1}^{(3)}} - e^{-i\omega_{1,1}^{(4)}} + \delta_{l} e^{-i\omega_{l}^{(2)}} \right] |1_{l}\rangle |0\rangle^{\otimes(n+1)}.$$ (21)

Here $\omega_{k}^{(1,2,3,4)}$ are the eigenfrequencies of the effective two-level system, and $\omega'_{l} = \omega_{l} - i\gamma'/2$.

In the following we consider two NV center case, in which we can quantify the entanglement using concurrence defined by Wootters [46]

$$C = \max \left\{ 0, \lambda_{1} - \lambda_{2} - \lambda_{3} - \lambda_{4} \right\},$$

(22)

where $\lambda_{1,2,3,4}$ are the square roots of the eigenvalues of the non-Hermitian matrix $\rho_{\sigma} \tilde{\rho}_{\sigma}$ [47], and $\tilde{\rho}_{\sigma}$ is the reduced density operator for the two qubits, $\tilde{\rho}_{\sigma} = (\sigma_{r} \otimes \sigma_{r})(\rho_{s}) \otimes \sigma_{r}$, with $\sigma_{r}$ being the Pauli operator. Under the initial state $|1\rangle |0\rangle |0_{\text{ccw}}\rangle |0_{\text{cw}}\rangle$ (i.e., one NV center is in |1⟩ state, the other is in |0⟩ state and the WGMs are in the vacuum state) we find $C = 2 |\rho_{12}|$. Further calculation shows that the concurrence is given by

$$C = \frac{1}{2} \left[ \begin{array}{c} \omega_{1,1}^{(3)} - \omega_{1,1}^{(4)} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right]^{2} - \left( \begin{array}{c} \omega_{1,1}^{(3)} - \omega_{1,1}^{(4)} \omega_{1,1}^{(1)} - \omega_{1,1}^{(2)} \end{array} \right)^{2}. $$

(23)

To obtain better entanglement between the two NV centers, the relative position of the two nanocrystals should be adjusted carefully, since the effective interaction between the two nanocrystals is sensitively dependent on their relative position.
For example, when \( \xi = \pi/2 + q \pi \), one nanocrystal only interacts with the ‘‘+’’ mode while the other nanocrystal only interacts with the ‘‘−’’ mode, which are independent of each other, yielding zero concurrence, as confirmed by Eq. (23). It is easy to verify that the concurrence is maximum for \( \xi = q \pi \), which corresponds to the totally constructive interference case, with the concurrence simplified as

\[
C = \frac{1}{2} e^{-\gamma t} \left| \frac{\Omega_+ + \Omega_-}{2 \Omega_0} - e^{-i \frac{\Omega_+ - \Omega_-}{2}} \right|^2 - 1,
\]

where \( \tilde{\Omega}_+ = \tilde{\Omega}_+^{(1)} + \tilde{\Omega}_+^{(2)} - 2 \omega_0' = - (\delta' + 4 \gamma') - i (\kappa_0/2 + 2 \kappa R - \gamma' / 2) \), \( \tilde{\Omega}_- = \tilde{\Omega}_-^{(1)} - \tilde{\Omega}_-^{(2)} = \sqrt{\Omega_0^2 + 16 G^2} \). Note that the parameter \( \tilde{\Omega}_+ \equiv \Omega_+ - i \Gamma_+ \) represents the total frequency shift (the real part \( \Omega_+ = - \delta' - 4 \gamma' \)) and total linewidth change (the imaginary part \( \Gamma_+ = \kappa_0 / 2 + 2 \kappa R - \gamma' / 2 \)) of the first and second eigenfrequencies, while the parameter \( \tilde{\Omega}_- \equiv \Omega_- - i \Gamma_- \) represents the difference of the two complex eigenfrequencies, in which the real part \( \Omega_- \) indicates the difference of the resonant position and the imaginary part \( \Gamma_- \) indicates the linewidth difference.

The entanglement between the qubits is affected not only by the relative positions of the nanocrystals, but also by the detunings and the nanocrystals’ size. According to Eq. (24), we plot the time evolution of the concurrence for various nanocrystals’ sizes in Fig. 5, and the maximum concurrence as a function of the two-photon detuning \( \delta \) and nanocrystals’ size \( R \) in Fig. 6. It is found that the entanglement properties are distinct for different nanocrystals’ sizes. We can divide the system into two regimes, the weak scattering regime and the strong scattering regime.

(i) Weak scattering regime. When the nanocrystals’ sizes are small, the scattering induced by the nanocrystals is weak. The frequency shift and linewidth broadening caused by the scattering can be neglected. When the decay rates of the microcavity and the effective two-level system are comparable (for example, \( 2 \kappa R + \kappa_0 / 2 + \gamma' / 2 \approx \gamma' \), this condition can be satisfied for relatively high-Q microcavity), the total linewidth change of the system can be neglected (i.e., \( \Gamma_+ \approx 0 \)). Then the concurrence can be simplified as

\[
C_1 \simeq e^{-\gamma t} \left| \frac{\Omega_+^2 - \Omega_-^2}{2 \Omega_0^2} \sin^2 \frac{\Omega_0 t}{2} \right| - i \left( \cos \frac{\Omega_0 t}{2} \sin \frac{\Omega_0 t}{2} - \Omega_+ \sin \frac{\Omega_0 t}{2} \cos \frac{\Omega_0 t}{2} \right).
\]

(25)

Here the concurrence has three oscillating components, with frequencies \( \Omega_+ \pm \Omega_- / 2 \) and \( \Omega_0 t \).

When the total frequency shift can also be neglected (i.e., \( \Omega_+ \simeq 0 \)), which corresponds to the resonant case (taking into account the light-induced frequency shift), we obtain \( \Omega_- = 4 |G'| \), and the concurrence can be simplified as

\[
C_1^{(1)} \simeq 2 e^{-\gamma t} \sin^2 (2 G' t).
\]

(26)

where only one oscillating component with frequency \( \Omega_- \) does not vanish. It is found that the concurrence cannot exceed 0.5. This is because the resonant coupling of the cavity mode and NV centers makes the cavity modes occupy the one-photon state with a large probability. This corresponds to the black solid lines in Fig. 5 and the blue areas in Figs. 6(a) and 6(c).

The large detuning case, \( |\Omega_-| \simeq |\Omega_+| \), from Eq. (25) we can see that only the oscillating component with frequency \( |\Omega_- - | / 2 \approx 4G' / |\Omega_+| \) exists, and the concurrence can be written as

\[
C_1^{(2)} \simeq e^{-\gamma t} \left| \sin \left( \frac{4G^2}{\Omega_0 t} \right) \right|.
\]

(27)

In this condition the concurrence can reach a value of near 1. The reason is that the NV centers interact dispersively with the cavity modes, thus the cavity modes are virtually excited. This corresponds to region II in Figs. 6(a) and 6(c).
The concurrence can also approach 1 in some “matched” conditions. When \( |\Omega_-| = (1 + q^{-1})|\Omega_+| \) (\( q = 1, 3, 5, \ldots \)), the frequencies \( |\Omega_\pm \pm \Omega_-|/2 \) and \( |\Omega_-| \) match with each other, which leads to a peak in the concurrence evolution. In these conditions the concurrence evolution has the maximum oscillation amplitude, apart from the \( e^{-\gamma t} \) decay envelope. This explains the fluctuations in region III of Fig. 6(a). Especially, \( |\Omega_-| \approx |\Omega_+| \) corresponds to \( |\Omega_-| = (1 + q^{-1})|\Omega_+| \) with \( q \) approaching infinity. For another example, \( |\Omega_-| = 2|\Omega_+| \), we obtain

\[
C_1^{(3)} \approx e^{-\gamma t} \frac{3}{8} \sin^2 \frac{4G'}{\sqrt{3}} t + i \sin^3 \frac{2G'}{\sqrt{3}} t. \tag{28}\]

The concurrence reaches its maximum when \( t \approx \sqrt{3}\pi/|4G'| \) and becomes zero when \( t \approx \sqrt{3}\pi/|2G'| \).

From Eqs. (27) and (28) it is found that when the concurrence reaches the maximum we obtain a mixed state of the maximum entangled state \((|\rangle|0\rangle \pm i|0\rangle|1\rangle)/\sqrt{2}\) of the two NV centers and the ground state.

(ii) Strong scattering regime. When the sizes of the nanocrystals are large, we arrive at the strong scattering regime. In the case that \( |\Omega_\pm|^2 \gg 4G^2 \), the concurrence can be simplified as

\[
C_2 \approx e^{-\left(\tilde{k}_\pm \pm \gamma'\right)t} |i \sin \left(\tilde{k}_\Omega \pm t\right) + \sinh \left(\tilde{k}_\Gamma \pm t\right)|, \tag{29}\]

where \( \tilde{k} = 4G^2/|\Omega_\pm|^2 \).

In the “resonant” case \( (\Omega_+ = 0) \), the \( \sinh(\tilde{k}_\Omega \pm t) \) term in Eq. (29) vanishes, and we obtain

\[
C_2^{(1)} \approx \frac{1}{2} e^{-\gamma' t} \left(1 - e^{-\tilde{k}_\Gamma \pm t}\right). \tag{30}\]

Note that \( C_{\text{max}} \) is not larger than 0.5. In the time evolution of concurrence, only a single peak appears and no oscillation exists. There are also no fluctuations in the maximum concurrence as the detuning changes.

For large detuning with \( |\Omega_\pm| \gg |\Gamma_\pm| \), the \( \sinh(\tilde{k}_\Gamma \pm t) \) term in Eq. (29) vanishes, and the concurrence can be simplified as

\[
C_2^{(2)} \approx e^{-\left(\tilde{k}_\pm \pm \gamma'\right)t} |\sin(\tilde{k}_\Omega \pm t)|. \tag{30}\]

We find that a large \( \Omega_\pm \), which can be realized by large detuning \( \Delta' \) or large scattering-induced coupling strength \( g \), will lead to large concurrence. Therefore, the stronger nanocrystal scattering will lead to better entanglement between the two NV centers for a fixed detuning.

For large nanocrystals’ size increases, the oscillation frequency becomes larger, and the maximum concurrence can exceed 0.5 (see red dashed line, \( R = 20 \) nm). For large nanocrystals’ size (\( R = 40 \) nm), a single peak appears and the maximum concurrence is still relatively large.

**VI. CONCLUSION**

In summary, we have established a model for the interaction between multiple diamond nanocrystals with single NV centers and WGMs of a microcavity. We calculate the eigenmodes and eigenfrequencies of the system and find that the system generally has four resonant frequencies, with degeneracies for some special relative positions of the nanocrystals. We present the transmission spectrum of the system with fiber taper coupling. It is found that the transmission spectrum exhibits a twofold-splitting phenomenon, where the first fold of splitting originates from the Rayleigh scattering of the nanocrystals and the second fold of splitting stems from the NV center dipole coupling with the WGMs. We study the entanglement of two NV centers via simultaneous coupling to the WGMs. The analytical results for the concurrence of two NV centers are presented. According to our analytical results, we find that the Rayleigh scattering of the nanocrystals can play a constructive role in generating entanglement between two NV centers when the detuning is fixed. We conclude that a large scattering strength is beneficial for obtaining a large concurrence for the fixed dipole-cavity detuning. It reveals that despite of the energy decay caused by the nanocrystals’ scattering, we can harness the scattering to improve the quantum properties of the system. This may open up promising perspectives for solid-state quantum information and quantum computation.
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