Cavity QED treatment of interactions between a metal nanoparticle and a dipole emitter

Edo Waks and Deepak Sridharan

Institute for Research in Electronics and Applied Physics, Joint Quantum Institute, University of Maryland College Park, Maryland 20742, USA
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We derive a full quantum optical model of interactions between a dipole and a metal nanoparticle. The electromagnetic field of the nanoparticle is quantized from the time-harmonic solution to the wave equation. We derive an analytical expression for the dipole-field coupling strength and the Purcell factor. The semiclassical theory, derived from the Maxwell-Bloch equations, is compared to the full quantum calculations based on numerical solution of the master equation. The metal nanoparticle-dipole system is found to be in an interesting regime of cavity quantum electrodynamics where dipole decay is dominated by dephasing, but the dipole-field coupling strength is still strong enough to achieve large cooperativity. In the presence of large dephasing, we show that simple semiclassical theory fails to predict the correct scattered field spectrum even in the weak-field limit. We reconcile this discrepancy by applying the random-phase-jump approach to the cavity photon number instead of the dipole operator. We also investigate the quantum fluctuations of the scattered field and show that they are significantly dependent on the dephasing rate.

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I. INTRODUCTION

The interactions between atomic systems and plasmonic structures is a topic of great interest for a broad range of applications in photonics and optoelectronics. Plasmonic particles exhibit strong electric field localization that can significantly enhance interactions with atomic media. This enhancement has been proposed and demonstrated as a mechanism for improving sensing capabilities via methods such as surface-enhanced Raman scattering [1]. It has also been investigated for increasing atomic radiative efficiency [2], guiding quantum dot excitation over nanowires [3,4], and nanoscale transistors [5].

To date there have been a large number of theoretical studies of interactions between dipoles and confined plasmonic structures. Studies of the radiative decay rates and quenching of dipole emitters near nanoparticles and surfaces have been reported in a number of works [2,6–11]. More recently, several works have shown that interactions between a dipole and a metal nanoparticle (MNP) can lead to interesting interference effects [12–16], resulting in sharp antiresonances or Fano line shapes of the scattering spectrum. These interference effects have been studied exclusively in the context of a semiclassical treatment, in which the dipole is treated as a quantum-mechanical two-level system but the MNP field is treated using classical mean-field theory. In addition, some works have begun to investigate the nonlinear behavior of this coupled system under strong-field excitation [12,16]. The semiclassical treatment is generally believed to be correct in the weak-field limit where the dipole is interacting with photons one at a time [17–19]. But it is known to be less reliable in the strong-field regime, where quantum fluctuations can influence, and sometimes wash out, nonlinear behavior such as optical bistability [20].

In this article we derive a model for the interaction between a dipole (a quantum-mechanical two-level system) and a spherical metal nanoparticle (MNP) using the principles of cavity quantum electrodynamics (cQED). In this treatment both the dipole and the electromagnetic field of the MNP are quantized, enabling a more precise analysis of their mutual interactions. For calculations we consider the specific case of a cadmium selenide (CdSe) quantum dot (QD) in proximity to a gold nanosphere. The cQED approach is found to be ideal for investigating the interference effects between an MNP and a dipole emitter in the presence of an external excitation field. Using this formalism an analytical expression for the dipole-MNP coupling strength and the Purcell factor is derived within the dipole approximation. The semiclassical Maxwell-Bloch equations are then determined directly from the fully quantized Heisenberg-Langevin equations. An analytical semiclassical expression for the scattered field and total absorbed power of the MNP is derived in the weak-field limit and shown to exhibit strong interference effects as predicted by previous semiclassical calculations. This interference is shown to be a consequence of the large atomic cooperativities that can be achieved in this system.

After performing the semiclassical analysis, we turn to the full quantum optical treatment performed by numerically solving the complete master equation. We show that in the strong-field limit, the interference between the MNP and QD are washed out due to QD saturation. In addition, we find that even in the weak-field limit the semiclassical approach fails to predict the absorbed power by more than an order of magnitude when dephasing is included. This failure is attributed to the mean-field approach which ignores the stochastic nature of the MNP field. We reconcile the discrepancy between the semiclassical and quantum theory by directly averaging the cavity photon number operator instead of the photon field. Finally, the quantum fluctuations of the scattered field are investigated and shown to exhibit a strong bunching effect which is highly dependent on the dephasing rate. In the limit of small dephasing the scattered field exhibits strong bunching in the weak-field limit. As the amount of dephasing is increased the degree of bunching is highly reduced.

The article is organized as follows: In Sec. II we quantize the electromagnetic field of an MNP and derive the basic Hamiltonian for interaction with a two-level quantum system.
In Sec. III we use the Heisenberg-Langevin formalism to derive an analytical expression for the Purcell factor and atomic cooperativity. The semiclassical Maxwell-Bloch equations are directly determined from this formalism and an analytical expression for the weak-field scattering spectrum is derived. Finally, in Sec. VI we provide a correct semiclassical calculation of the scattered field.

In Sec. IV we perform a fully quantum calculation through numerical simulation of the master equation. In Sec. V we consider two specific excitation directions, longitudinal and transverse. In longitudinal excitation the polarization of the light lies parallel to the line connecting the centers of the two nanoparticles, while in transverse excitation the polarization is orthogonal to this line. All intermediate excitation directions can be decomposed into a longitudinal and transverse component which are independent provided the MNP and dipole are spherically symmetric.

The quantization of an MNP has been investigated in a number of previous works in the context of radiative lifetime modification [2,6–11]. A quantity of central importance to these works is the dyadic Green function that relates the local electric field to the surface charge distribution of the MNP. Here we utilize a different but mathematically equivalent procedure that directly works with the electric field of the MNP. In this procedure we treat the MNP as an optical resonator with discrete optical modes. The modes of the resonator are identified by finding localized solutions to Maxwell’s equations subject to the boundary conditions of the metal dielectric interface. For each of these localized modes we find a pair of canonical conjugate variables that directly lead to quantized field operators. This approach is identical to field quantization of more standard resonators such Fabry-Perot cavities [21], with a few additional subtleties due to the subwavelength nature and negative dielectric constant of the MNP. These subtleties will be addressed when they come up.

To quantize the MNP field, we begin with its classical electromagnetic description. The dielectric constant of the metal is given by $\epsilon_1$, while the dielectric constant outside the MNP is given by $\epsilon_2$. In some cases it is advantageous to encase the MNP in a dielectric material other than vacuum in order to shift its plasmon resonance. We assume the MNP is nonmagnetic so the permeability is given by $\mu = \mu_0$. The size of the MNP is assumed to be small compared to the wavelength of light so that we can make the time harmonic approximation that the electric field can be written as $E(r,t) = \text{Re}[E(r)e^{-i\omega t}]$, where $E(r)$ is the electric field associated with the solution of the Laplace equation for the electrostatic scalar potential, and Re{} indicates the real part of the bracketed quantity. If the MNP is driven by an external monochromatic field $E_0(r,t) = \text{Re}[E_0e^{-i\omega t}]$, where $E_0$ is a constant vector, then the total field which includes the response of the MNP is given by

$$E(r) = E_0 + \sum_{i=x,y,z} E_{0i} \frac{\epsilon_2 - \epsilon_1}{2\epsilon_2 + \epsilon_1} G_i(r),$$

where $i$ sums over the three-dimensional coordinates, $E_{0i}$ are the components of $E_0$, and

$$G_i(r) = \begin{cases} \hat{i} & r < R \\ -\frac{r^i}{r^3}[3(\hat{\mathbf{r}} \cdot \hat{i})\hat{\mathbf{r}} - \hat{i}] & r > R \end{cases}$$

In the above equation $R$ is the radius of the MNP, $r$ is the radial coordinate of the position vector $\mathbf{r}$ (taken with respect to the center of the MNP), and $\hat{\mathbf{r}}$ is a unit vector pointing in the direction of $\mathbf{r}$. The unit vector $\hat{i}$ points in the direction of the $i$th coordinate axis. From Eq. (1) it is clear that the electric field can be written as a sum of the externally applied electric field plus the electric field of the MNP given by

$$E_{\text{mnp}}(r) = \sum_i E_i(r),$$

where

$$E_i(r) = E_{0i} \frac{\epsilon_2 - \epsilon_1}{2\epsilon_2 + \epsilon_1} G_i(r).$$

We note that the above fields satisfy orthogonality in that

$$\int E_i(r) \cdot E_j(r) d^3r = 0$$

if $i \neq j$.

Because the MNP is metallic, the dielectric constant $\epsilon_1$ can be well modeled by a Drude dispersion relation given by

$$\epsilon_1 = 1 - \frac{\omega_p^2}{\omega(\omega - i\gamma_o)},$$

where $\omega_p$ is the plasma frequency of the metal and $\gamma_o$ accounts for energy dissipation due to ohmic losses in the medium.
Appendix A we show that when $\gamma_0 < \omega$ the field of the MNP can be approximated as
\begin{equation}
E_i(r, \omega) \approx E_0 |G_i(r)| \eta \frac{\omega_0/2}{(\omega - \omega_0) - i\gamma_0/2},
\end{equation}
where $\eta = 3\epsilon_2/(2\epsilon_2 + 1)$ and $\omega_0 = \omega_p/\sqrt{2\epsilon_2 + 1}$. In this limit, the electric field response follows a Lorentzian spectrum. The Lorentzian approximation to the MNP spectral response is expected to be very good when it is interacting with a dipole whose resonant frequency is close to plasmon resonance, which is the primary regime of interest for this article.

To quantize the MNP field we need to find localized solutions to Maxwell’s equations. These solutions are bounded in space and decay to zero far away from the MNP. The solution presented previously is not a localized solution because it is presented in the time domain is
\begin{equation}
G = \frac{2\omega_0}{\omega(\omega - \omega_0) - i\gamma_0/2},
\end{equation}
is initially ignored and the field must be incorporated properly by coupling the quantized mode with a continuum of reservoir modes. This approach ensures that the quantized field operators obey the same commutation relations for all time. From the above equation one can see that the $G_i(r)$ provides the electric field distribution for the localized mode. One can readily verify that $G_i(r)$ satisfies both Maxwell’s equations (in the time harmonic limit) and the boundary conditions at frequency $\omega = \omega_0$ in the undamped limit. It therefore represents a localized solution to Maxwell’s equations.

Because the three modes of the MNP satisfy orthogonality, we may quantize each one of them individually. The energy in the electric field of the $i$th mode is given by
\begin{equation}
W_i = \alpha_i^2 \frac{\epsilon_0}{2} \sin^2 \omega_0 t \int |G_i(r)|^2 \text{Re} \left[ \frac{d}{d\omega} \omega(e^{i\omega t} |_{\omega = \omega_0}) \right] d^3r.
\end{equation}
We note that energy in the above equation has to be defined properly using the more precise expression for dispersive materials [22] because the MNP has a negative dielectric constant. If we use the nondispersive expression it leads to negative energies that will result in un-physical negative mode volumes. We next define a normalized variable $A_{i0}$ given by
\begin{equation}
A_{i0} = \frac{\alpha_i}{N},
\end{equation}
where
\begin{equation}
N = \sqrt{\frac{2\hbar \omega_0}{\epsilon_0 \int |G_i(r)|^2 \text{Re} \left[ \frac{d}{d\omega} \omega(e^{i\omega t} |_{\omega = \omega_0}) \right] d^3r}.\end{equation}
Using this normalized amplitude we can write
\begin{equation}
W_i = \hbar \omega_0 A_{i0}^2 \sin^2 \omega_0 t.
\end{equation}
The above expression represents the stored energy in the electric field. In most optical resonators field quantization would involve adding the energy stored in the magnetic field to form the Hamiltonian. The electric and magnetic fields would then form a pair of canonical conjugate variables that satisfy a Hamiltonian for a quantum harmonic oscillator. However, because we are quantizing the near-field mode of the MNP, the magnetic field is zero because the electric field can be written as the gradient of a scalar potential.

To resolve this problem, we note that for energy conservation to be preserved the total energy of the system must be constant. This energy is continually being converted from stored potential energy, represented by the energy of the field, and kinetic energy due to current flowing in the MNP. To maintain energy conservation, we must add a second term in the Hamiltonian to account for the kinetic energy of the current, and this term must be of the form
\begin{equation}
K_i = \hbar \omega_0 A_{i0}^2 \cos^2 \omega_0 t,
\end{equation}
in order for the total energy of the system to be constant. In this way the MNP is identical to a normal harmonic oscillator where the energy is periodically converted between the kinetic and potential energy. The total Hamiltonian is attained by adding these two components to give $H_i = W_i + K_i = \hbar \omega_0 A_{i0}^2$ at all times. We can now write
\begin{equation}
A_i(t) = A_{i0} \sin \omega_0 t.
\end{equation}
Using this definition we can write the Hamiltonian as
\begin{equation}
H_i = \frac{\hbar}{\omega_0} (\omega_0^2 A_i^2 + \dot{A}_i^2),
\end{equation}
where $\dot{A}_i$ is the time derivative of $A_i$. The above is a Hamiltonian for a harmonic oscillator, which can be easily seen by substituting $\hbar/\omega_0 \rightarrow m/2$. This substitution leads to the standard Hamiltonian for a mechanical oscillator with mass $m$ and resonant frequency $\omega_0$. The two variables $A_i$ and $2\hbar \dot{A}_i/\omega$ form a pair of canonical conjugate variables that can be quantized.

To quantize the MNP field we first transform the two scalar conjugate variables into quantum operators as $A_i \rightarrow \hat{x}_i$ and $2\hbar \dot{A}_i/\omega \rightarrow \hat{p}_i$, which satisfy the commutation relation
\begin{equation}
[\hat{x}_i, \hat{p}_i] = i\hbar.
\end{equation}
We then define the bosonic field operator $\hat{a}_i$ as
\begin{equation}
\hat{a}_i = \hat{x}_i + \frac{i}{2\hbar} \hat{p}_i.
\end{equation}
Using this definition the Hamiltonian takes on a simpler form given by
\begin{equation}
H_{\text{mp}} = \hbar \omega_0 \sum_{i=x,y,z} (\hat{a}_i^\dagger \hat{a}_i + 1/2).
\end{equation}
From Eq. (16) it is straightforward to show that
\begin{equation}
[\hat{a}_i, \hat{a}_i^\dagger] = 1.
\end{equation}
Equations (10) and (14) can be combined to write
\[ \alpha_i \rightarrow \frac{N}{2} (\hat{\alpha}_i + \hat{\alpha}_i^\dagger). \tag{20} \]

Inserting this expression into Eq. (8) we attain the quantized electric field operator
\[ \hat{E}_{\text{mnp}} = \sqrt{\frac{\hbar \omega_0}{2 \epsilon_0 V_m}} \sum_i Y_i(r) (\hat{\alpha}_i + \hat{\alpha}_i^\dagger), \tag{21} \]
where the mode volume \( V_m \) is defined as the ratio of the total energy to the energy density inside the MNP, given by
\[ V_m = \int |G_i(r)|^2 \frac{d^3 r}{\omega \epsilon} \frac{d^3 r}{\omega \epsilon} \frac{\omega \epsilon}{2} \tag{22} \]
and
\[ Y_i(r) = \frac{G_i(r) \sqrt{\omega \epsilon}}{\left|G_i(0)\right|^2 \frac{d^3 r}{\omega \epsilon}} \frac{\omega \epsilon}{2 \sqrt{(2 \epsilon_2 + 1)}} \tag{23} \]
The function \( Y_i(r) \) is a rescaled version of \( G_i(r) \). The function \( G_i(r) \) is normalized such that the electric field inside the MNP is unity, while the function \( Y_i(r) \) is normalized such that it gives an energy density of unity instead. We note that our definition of the mode volume differs slightly from the usual definition for a dielectric resonator where one normalizes by the peak value of the energy density which, for the MNP, would be achieved at the surface. We choose this slightly different definition because it leads to a more intuitive expression for the mode volume. From Eq. (2) the mode volume can be calculated in a straightforward way to give
\[ V_m = \frac{4}{3} \pi R \frac{(2 \epsilon_2 + 1)}{(\epsilon_2 + 1)}. \tag{24} \]

Having attained the quantized electric field operator, we can now define the interaction Hamiltonian for the MNP with the dipole. We assume that this interaction is dipolar in nature and is therefore given by \( H_{\text{int}} = -\hat{q} \cdot \hat{E}_{\text{mnp}} \), where \( \hat{q} \) is the dipole operator. The validity of the dipole approximation has been investigated in previous works [2,15] that have shown that when the distance between the MNP and the dipole is larger than the radius of the MNP the dipolar approximation provides a reasonable estimate for the interaction strength. The effect of higher-order multipole contributions has been investigated semiclassically [15] and become particularly important when the spacing between the particles is less than the MNP radius. The dipole operator can in general be written as
\[ \hat{q} = \sum_{mn} q_{mn} \sigma_{mn}, \tag{25} \]
where \( q_{mn} \) are the matrix elements of the dipole operator between the \( m \)th and \( n \)th energy eigenstates and \( \sigma_{mn} = |n \rangle \langle m| \).

For a spherically symmetric dipole we can always select a quantization direction such that \( q_{mn} \) points along the \( x \), \( y \), or \( z \) axis. The interaction Hamiltonian will couple field operators only with dipole operators that are pointing in the same direction. If we excite only one specific transition of the dipole so that we can make the two-level dipole approximation, then the dipole will only couple to one of the modes of the MNP. In this limit, we can treat the dipole and MNP as a two-level system interacting with a single bosonic mode whose field is either in the longitudinal or transverse direction. The case where multiple modes of the dipole are excited can be handled in a straightforward way because each transition will couple only to one mode of the MNP. Situations where the MNP does not have spherical symmetry, i.e., a nonradon, can also be handled analogously but the coupling between the MNP and dipole will now depend on the orientation of the MNP. In the asymmetric case it is also possible for a transition of the dipole to excite more than one of the MNP electromagnetic modes. We will not consider these more general situations here.

Making the two-level approximation, the dipole operator can be written in the form \( \hat{q} = \mu \hat{k} (\sigma_- + \sigma_+) \), where \( \mu \) is the dipole moment of the transition and \( \sigma_- \) and \( \sigma_+ \) are the Pauli raising and lowering operators respectively. We define \( \hat{a} \) as the bosonic operator corresponding to the single MNP mode interacting with the dipole. We apply the rotating wave approximation and drop the energy nonconserving terms, which leads to the standard Jaynes-Cummings Hamiltonian for a two-level system interacting with a single electromagnetic mode, given by
\[ H_{\text{int}} = \hbar g(r)(\sigma_+ \hat{a} + \hat{a}^\dagger \sigma_-), \tag{26} \]
where \( g(r) \) is the MNP-dipole coupling strength or, alternately, the vacuum Rabi frequency. This term is given by
\[ g(r) = \begin{cases} -2 \frac{\mu}{\hbar} \sqrt{\frac{m_{\text{mp}}}{2 \omega_0}} R^3 & \text{longitudinal coupling} \\ -\frac{\mu}{\hbar} \sqrt{\frac{m_{\text{mp}}}{2 \omega_0}} R^3 & \text{transverse coupling}. \end{cases} \tag{27} \]

We note that the vacuum Rabi frequency is twice as strong for longitudinal excitation, indicating that this coupling configuration is preferable for strong interactions.

We have quantized the field of the MNP and have properly derived the interaction Hamiltonian between the MNP and the dipole. By doing so, we can now analyze the system using the vast body of analytical and computational techniques of cavity quantum electrodynamics. These approaches can provide tremendous insight into this problem because they typically characterize the complex physics of atom-light interaction with only a handful of parameters.

### III. HEISENBERG-LANGEVIN ANALYSIS

The Heisenberg-Langevin approach gives a physically intuitive picture of the interaction between an optical field and a dipole because it provides a set of operator equations that mirror the semiclassical Maxwell-Bloch equations. This approach also provides a simple method for handling damping and excitation of the system. Damping can be handled by open system theory, in which the MNP is coupled to a continuum of damping modes. MNPs can be damped both nonradiatively due to ohmic losses and radiatively due to dipole emission. We define \( \hat{b}_\omega \) as the continuum of radiative output modes and \( \hat{c}_\omega \) as a reservoir of phonon modes that can carry energy away due to heating. The Hamiltonian for the reservoir modes is given by Refs. [21,23,24]
\[ H_{\text{res}} = \int \hbar \omega'(\hat{b}_\omega^\dagger \hat{b}_\omega + \hat{c}_\omega^\dagger \hat{c}_\omega) \ d\omega' \]
and the system reservoir interaction is given by
\[ H_{\text{int}} = i \hbar \int \left[ T_1 b_{\text{ad}}^\dagger \hat{a} + T_2 \hat{e}_{\text{ad}}^\dagger \hat{a} + T_3 b_{\text{ad}}^\dagger \sigma_- \right] d\omega' + \text{H.c.} \] (29)

In the above equation \( T_1 \) and \( T_2 \) represent the coupling strength between the MNP and the reservoir modes while \( T_3 \) represents the coupling strength between the dipole and the radiative modes. We note that, because the distance between the dipole and the MNP is much smaller than the wavelength of light, there is no phase retardation between their relative interactions with the radiative modes. In addition to damping, we need to handle the excitation of the MNP and dipole due to an external field. We assume that both the MNP and dipole are driven by a classical monochromatic field (or equivalently a single mode coherent state). The interaction between the excitation field and system is given by the Hamiltonian [20,24]
\[ H_{\text{drive}} = \frac{\hbar}{2} \kappa (\hat{a} e^{-i\omega t} - \hat{a}^\dagger e^{i\omega t}) + i\hbar (\sigma_+ \Omega e^{-i\omega t} - \sigma_- \Omega e^{i\omega t}), \] (30)
where \( \kappa \) is the decay rate of the MNP (to be derived from the reservoir interactions) and \( \omega \) is the driving frequency of the external field. The parameter \( \epsilon \) is the driving amplitude for the MNP, while \( \Omega \) is the classical Rabi frequency of the external field driving the dipole. Both \( \epsilon \) and \( \Omega \) are proportional to the incident electric field amplitude \( E_0 \). We will derive this relationship explicitly but to do so we must first derive the Heisenberg equations of motion.

We begin with the Hamiltonian of the combined MNP-dipole system given by
\[ H = H_{\text{MNP}} + H_{\text{dip}} + H_{\text{int}} + H_{\text{res}} + H_{\text{scat}} + H_{\text{drive}}, \] (31)
where \( H_{\text{MNP}} \) is given by Eq. (18) and \( H_{\text{int}} \) is given by Eq. (26). The system Hamiltonian for the dipole is given by
\[ H_{\text{dip}} = \frac{\hbar \omega_d}{2} \sigma_z, \] (32)
where \( \omega_d \) is the resonant frequency of the dipole and \( \sigma_z \) the Pauli operator for the population inversion. Using this Hamiltonian we can derive the Heisenberg equations of motion for all the operators. In Appendix B we show that the above Hamiltonian directly leads to the Heisenberg-Langevin equations of motion given by
\[ \frac{d\hat{a}}{dt} = -(i\Delta_+ + \kappa/2)\hat{a} - ig\sigma_- + \sqrt{\kappa} \hat{f} + \hat{g}, \] (33)
\[ \frac{d\sigma_-}{dt} = -(i\Delta_- + \gamma_0/2)\sigma_- + ig\sigma_z \hat{a} - \Omega \sigma_z + \hat{g}, \] (34)
\[ \frac{d\sigma_z}{dt} = -\gamma_0(\sigma_z + I) + 2ig(\hat{a}^\dagger \sigma_- - \sigma_- \hat{a}) + 2\Omega(\sigma_- + \sigma_+) + \hat{f}. \] (35)

In the above equations \( \gamma_0 \) is the spontaneous emission rate of the dipole and \( \kappa = \gamma_0 + \gamma_r \) is the total decay rate of the MNP due to both ohmic losses in the metal (\( \gamma_0 \)) and scattering into free-space modes (\( \gamma_r \)). The scattering rate \( \gamma_r \) can be calculated classically from the Larmor formula. We show in Appendix C that for a metal sphere \( \gamma_r = 2\omega_0^3\epsilon_0 R^3/c^3(2\epsilon_2 + 1) \). The cavity and dipole detunings are given by \( \Delta_+ = \omega_d - \omega \) and \( \Delta_- = \omega_0 - \omega \). The operators \( \hat{f}, \hat{g}, \hat{h}, \) are the noise operators that conserve the commutation relations at all times.

In many cases we will be interested in the total amount of power absorbed by the MNP. This quantity can be calculated using input-output formalism [23] by defining the output flux operator
\[ \hat{b}_{\text{out}}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \hat{b}_{\text{in}}(t') e^{-i\omega(t-t')} d\omega', \] (36)
where \( t_f \) is a final time long after all interactions have ended. In Appendix D we derive the relationship between the output flux and the MNP and dipole operator, which is given by
\[ \hat{b}_{\text{out}} = \sqrt{\gamma_r} \hat{a} + \sqrt{\gamma_s} \sigma_- \] (37)
The above expression extends the standard cavity input-output relation given in Ref. [23] to the case where the dipole can also excite the external field modes. The average rate at which energy is scattered into free-space modes is given by \( \Phi_{\text{scat}} = \langle \hat{b}_{\text{out}} \hat{b}_{\text{out}}^\dagger \rangle \), which can be written as
\[ \Phi_{\text{scat}} = \gamma_r (\hat{a}^\dagger \hat{a}) + \gamma_d \rho_{22} + \sqrt{\gamma_r} \sqrt{\gamma_s} (\hat{a} \sigma_- + \sigma_+ \hat{a}), \] (38)
where \( \rho_{22} \) is the excited state probability of the dipole (or alternate the diagonal element of the dipole reduced density matrix). Using the identical mathematical formalism we can derive an expression for the energy flux absorbed in the form of phonons which is given by the output relation
\[ \hat{e}_{\text{out}} = \sqrt{\gamma_r} \hat{a}. \] (39)
The energy dissipation rate due to phonons is then given by
\[ \Phi_{\text{dis}} = \gamma_0 (\hat{a} \hat{a}) \] (40)
The total absorbed power can be calculated by summing both dissipation mechanisms to give \( \Phi_{\text{abs}} = \Phi_{\text{dis}} + \Phi_{\text{scat}} \).

Having derived the Heisenberg-Langevin equations, we can now provide a physical interpretation for the classical driving amplitude \( \epsilon \). To do this we consider the case of a bare MNP that does not contain a QD (\( g = 0 \)) and is excited on resonance (\( \Delta_+ = 0 \)). We take the expectation value of both sides of Eq. (33), define \( A = \langle \hat{a} \rangle \), and solve for the steady-state solution of the field amplitude to give
\[ A = \frac{2\epsilon}{\sqrt{\kappa}}. \] (41)
The above is derived using the fact that the output modes are initially in vacuum so the \( \langle \hat{f} \rangle = 0 \). Using this expression the total power absorbed by the MNP is then given by \( \Phi_{\text{abs}} = \Phi_{\text{scat}} + \Phi_{\text{dis}} = 4|\epsilon|^2 \). Thus, \( 4|\epsilon|^2 \) is the rate of energy absorption in units of photons per second. By taking the expectation value of Eq. (21), and remembering that \( A \) was derived in the rotating frame and must be multiplied by \( e^{-i\omega t} \) to go back to the laboratory frame, we attain the expression
\[ \langle \hat{E}_{\text{MNP}} \rangle = \sqrt{\frac{\hbar \omega_0}{4e_0 V_{\text{A}}(\epsilon_2 + 1)}} \text{Re} \left\{ G(r) \frac{2\epsilon}{\sqrt{\kappa}} e^{-i\omega t} \right\}. \] (42)
The classically derived equation for the MNP field amplitude driven on resonance by a monochromatic field is given by Eq. (7). Substituting \( \gamma_0 \) with \( \kappa \) so that we account for both radiative and nonradiative decay we then have the classical expression
\[ E_{\text{MNP}} = E_0 \text{Re} \left\{ iG(r) \frac{\omega_0}{\kappa} e^{-i\omega t} \right\} \] (43)
By setting the classically derived equation equal to the quantum mechanically derived equation we obtain the relationship between \( E_0 \) and \( \epsilon \) given by

\[
E_0 = -i \frac{\epsilon}{\eta} \sqrt{\frac{\hbar \kappa}{\omega_0 \epsilon_0 V_m (\epsilon_2 + 1)}}. \tag{44}
\]

This expression allows us to convert between the normalized driving amplitude \( \epsilon \) and the electric field amplitude of the incident electromagnetic wave. The Rabi frequency \( \Omega \) can be derived in terms of \( \epsilon \) using the well-known relationship \( \Omega = i E_0 \mu / 2 \hbar \). Thus, for all of our calculations we can specify \( \epsilon \) only, we do not need to separately specify \( \Omega \) as both of these terms originate from the same driving source \( E_0 \).

For the majority of practical dipole emitters the MNP damping rate \( \kappa \) is many orders of magnitude larger than the dipole spontaneous emission rate \( \gamma_s \). In this limit we can adiabatically eliminate \( \hat{a} \) from Eq. (35) by substituting the steady-state solution to Eq. (33) given by

\[
\hat{a} = \frac{\sqrt{\kappa \epsilon}}{i \Delta_c + \kappa / 2} \hat{\sigma}_+ + \hat{\mathbf{f}}, \tag{45}
\]

which leads to the new equation of motion for \( \sigma_z \) given by

\[
\frac{d\sigma_z}{dt} = -\Gamma (\sigma_z + I) + 2 (\Omega \sigma_+ + \Omega^* \sigma_-) + \hat{\mathbf{f}}', \tag{46}
\]

where

\[
\Omega_i = \Omega - i \frac{\sqrt{\kappa \epsilon}}{i \Delta_c + \kappa / 2}, \tag{47}
\]

while the new noise operator is given by

\[
\hat{\mathbf{f}}' = \hat{\mathbf{h}} + 2i g \left( \hat{\mathbf{f}} \hat{\sigma}_- - \hat{\sigma}_- \hat{\mathbf{f}} \right) \tag{48}\]

and the modified spontaneous emission rate of the dipole is given by

\[
\Gamma = \gamma_s + \frac{4 g^2 \kappa}{\kappa^2 + 4(\omega_0 - \omega_a)^2}. \tag{49}
\]

The above expression is identical to previously derived decay rates based on Green’s function formalism [11] but is now derived in the context of a cQED problem. We define the Purcell factor as the ratio of the modified to unmodified spontaneous emission rate, which is given by

\[
F_p = 1 + \frac{4 g^2 \kappa}{\gamma_s (\kappa^2 + 4 \delta^2)}. \tag{50}
\]

Note that in the above expression we have assumed that the MNP does not modify the dipoles ability to radiate into the additional vacuum modes that exist in free space. Thus, \( \gamma_s \) is assumed to be given by the standard Wigner-Weisskopf spontaneous emission rate of the dipole.

To calculate the Purcell factors achievable in this system we focus on the specific example of a cadmium selenide (CdSe) quantum dot interacting with a metallic gold nanosphere. The values of \( \omega_0 \) and \( \gamma_s \) for the MNP are determined by first calculating the exact response of the gold nanosphere using numerical values for the dielectric constant of gold taken from the article of Johnson and Christy [25]. The sphere is assumed to have a radius of \( R = 5 \text{ nm} \) and be embedded in a material with an index of \( \epsilon_2 = 1.5 \). In practice this condition can be attained by capping the system with a polymer layer such as poly(methyl methacrylate) (PMMA). Encapsulation of both CdSe QDs and gold nanospheres in PMMA has been experimentally demonstrated in a number of works [26,27]. We fit the calculated response to a Lorentzian function, and from the fit we determine \( \gamma_0 = 2 \pi c / \omega_0 = 550 \text{ nm} \), which is a compatible wavelength for interactions with CdSe nanocrystals. The quality factor of the MNP due to ohmic losses is given by \( Q_d = \omega_0 / \gamma_0 = 11 \), while the quality factor for scattering is given by \( Q_s = \omega_0 / \gamma_s = 19000 \), and the total quality factor is attained by \( Q = (1/Q_d + 1/Q_s)^{-1} \). The decay rate is then calculated by \( \kappa = \omega_0 / Q \). We note that \( Q_r \) is significantly higher than \( Q_d \), thus we expect a significant fraction of absorbed power to be dissipated as heat due to ohmic losses rather than scattered. For the QD, we set \( 1/Q_r = 10 \text{ ns} \), corresponding to the radiative decay rate of a CdSe nanocrystal [27]. Figure 2 plots the calculated Purcell factor as a function of distance from the center of the MNP and the location of the dipole for both longitudinal and transverse coupling. The Purcell factor can become extremely large when the QD is within a few nanometers of the MNP and is larger for longitudinal coupling due to the enhanced value of the dipole coupling parameter.

We next use the Heisenberg-Langevin equations to derive the semiclassical Maxwell-Bloch equations, which enables us to rederive semiclassical results using the notation introduced from the full quantum optical formalism. To derive these equations we take the expectation values of both sides of Eqs. (33), (34), and (46). We take the cold reservoir limit where the reservoir modes are all initially in vacuum states. In this limit the reservoirs act only to draw away energy and do not excite the system via thermal excitations. This assumption is extremely good for optical frequencies and temperatures at or below room temperature. In the cold reservoir limit the expectation values of all noise operators can be neglected because they are annihilated when acting on the initial vacuum state. We define the expectations of the operators as \( \langle \sigma \rangle = A \), \( \langle \sigma \rangle = \rho_{21} \), and \( \langle \sigma \rangle = w \). In semiclassical formalism the field is a well-defined (i.e., noise free) classical amplitude so the expectations of products of a field operator and dipole operator are separable (i.e., \( \langle \sigma_+ \hat{a} \rangle = \langle \sigma_- \rangle \langle \hat{a} \rangle \)).
To this point we have still not included the effect of dephasing, which is typically handled by the random-phase-jump approach. The dipole is assumed to undergo unitary evolution until it experiences a dephasing event. Once a dephasing event occurs, the dipole moment averages out to zero, but the population inversion is unaffected. The average time scale over which a dephasing event occurs is defined as $T_2 = \gamma_d^{-1}$, where $T_2$ is the dipole coherence time. The probability that a dephasing event occurs on a time interval $dt$ is given by $p_d = dt \times \gamma_d$. The time evolution of the dipole term $\rho_{21}$ is then given by

$$\rho_{21}(t + dt) = \rho_{21}(t) + \frac{d\rho_{21}}{dt} dt - p_d \rho_{21}(t),$$

(51)

where the time derivative of $\rho_{21}$ accounts for evolution with no dephasing event which is given by taking the expectation value of Eq. (34). The portion of the matrix element that has undergone dephasing is subtracted off because it averages to zero. Using the above random-phase-jump model along with the semiclassical approximation we attain the Maxwell-Bloch equations for the MNP-dipole system which are given by

$$\frac{dA}{dt} = -(i\Delta_e + \kappa/2)A - ig\rho_{21} + \sqrt{\kappa} \rho_1,$$

(52)

$$\frac{d\rho_{21}}{dt} = -(i\Delta_d + \gamma_a)\rho_{21} + ig\omega A - \Omega w,$$

(53)

$$\frac{dw}{dt} = -\Gamma(w + 1) + 2(\Omega_d \sigma_+ + \Omega_c \sigma_-),$$

(54)

where $\gamma_a = \gamma_d/2 + \gamma_r$. For subsequent calculations, we set the dephasing rate to $\gamma_d = 7.6 \text{ ns}^{-1}$, which corresponds to the low temperature linewidth of 0.01 meV [28]. This linewidth is two orders of magnitude larger than the spontaneous emission rate.

The steady-state solution to Eq. (54) is

$$w = -1 + 4\text{Re} \left\{ \frac{\Omega_r \rho_{12}}{\Gamma} \right\}.\quad (55)$$

If $\Omega_r \ll \Gamma$ then $w \approx -1$. In this limit, which we refer to as the weak-field limit, a simple analytical solution can be attained for the MNP field and dipole moment. These solutions are given by

$$A = \frac{\sqrt{\kappa} \rho_1}{i\Delta_e + \gamma_a + \Gamma^2},$$

(56)

and

$$\rho_{21} = \frac{\Omega_r}{i\Delta_d + \gamma_a + \Gamma^2}.$$

(57)

If both the dipole and the field are resonant with the MNP the expression for the MNP field simplifies to $\Lambda = 2(\epsilon/\sqrt{\kappa} - ig\Omega/k\gamma_0)/(1 + C)$, where $C = 2g^2/\gamma_a\kappa$ is the atomic cooperativity. When $C \ll 1$ the cavity field is a sum of the response from the direct driving field and the driven dipole. If $C \gg 1$ the interaction between the dipole and the MNP will significantly suppress the cavity field.

Figure 3 plots the atomic cooperativity as a function of distance between the centers of the MNP and the QD for both transverse and longitudinal excitation using the same numbers used to calculate the Purcell factor. Longitudinal excitation achieves a cooperativity that is 4 times stronger due to the factor of two enhancement in the dipole coupling. One can see that atomic cooperativities greater than 1 are possible but only when the QD is within about 15 nm of the MNP. At larger distances the cooperativity falls off quickly due to the rapid decay of the electric field. It is rather remarkable that such cooperativities are possible despite the extremely low $Q$ of the MNP and large dephasing rate of the QD.

In Fig. 4 we plot the total absorbed power $\Phi_{\text{abs}} = \Phi_{\text{dis}} + \Phi_{\text{scat}}$ as a function of detuning. The absorbed power is normalized by $|A|^2$, which is the maximum absorbed power for a bare MNP. Calculations are performed for a longitudinal coupling configuration. The energy dissipated by ohmic losses is given in the semiclassical limit by $\Phi_{\text{dis}} = \gamma_a |A|^2$, while the scattered power is given by $\Phi_{\text{scat}} = \gamma_r |A|^2 + 2\sqrt{\gamma_r \gamma} \text{Re} \{\Omega_r \rho_{12} \} + \gamma_s \rho_{22}$. The excited state population $\rho_{22}$ can be determined from the relation $\rho_{22} = (w + 1)/2$. From Eq. (55) we then have

$$\rho_{22} = 2\text{Re} \left\{ \frac{\Omega_r \rho_{12}}{\Gamma} \right\},$$

(58)

which can be directly used to calculate the scattered power.

In Fig. 4(a) the distance between the MNP and QD is set to be $r = 10$ nm. At this distance the cooperativity is large ($C = 13.7$), leading to a strong antiresonance behavior where the absorption is significantly suppressed on resonance with the QD. The slight asymmetry is due to interference between the driving terms of the MNP and QD respectively. Figure 4(b) plots the absorption spectrum when $r = 25$ nm. In this case the atomic cooperativity is small ($C = 0.06$). However, the interference between the driving term of the MNP and the direct driving of the QD leads to a Fano line shape. These absorption spectra are consistent with previous semiclassical calculations [12–16]. The strong antiresonance behavior shown in Fig. 4(a) is seen to be the result of large cooperativity between the dipole and electromagnetic mode. Such interference has been previously studied in the context of an atomic system interacting with a single cavity mode both theoretically and experimentally [17–19,29–31].

IV. FULL QUANTUM CALCULATIONS THROUGH MASTER EQUATION APPROACH

In the previous section we calculated the absorption spectrum of the MNP in the semiclassical weak-field limit.
A natural extension of this analysis would be to consider the semiclassical model in the strong-field limit where we do not assume that \( w \to -1 \). This regime has been investigated in a number of works [20,32–35] and has been shown to exhibit interesting nonlinear effects such as optical bistability. The validity of the semiclassical analysis in the strong-field limit was investigated previously by Savage and Carmichael [20], who showed that when taking into account quantum fluctuations, the bistable states are in fact only metastable.

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FIG. 4. (Color online) Absorber power as a function of detuning from the MNP resonance when (a) the QD is located 10 nm from the MNP and (b) when the QD is located 25 nm from the MNP.

accounts for the damping of the MNP field and

\[
\mathcal{L}_{\text{dip}} \rho = -\frac{\gamma_s}{2}(\sigma_+ \rho \sigma_- - \rho \sigma_+ \sigma_-) - \frac{\gamma_d}{2}(\sigma_+ \rho \sigma_- + \rho \sigma_+ \sigma_- - 2\sigma_- \rho \sigma_+) \tag{62}
\]

accounts for spontaneous emission and dephasing of the dipole.

The Hilbert space of the MNP is in general infinite because it spans all photon number states. To perform calculations we must truncate the Hilbert space at some finite photon number states is negligibly small. For all calculations performed in this article the Hilbert space was truncated at 10 photons. Simulations were performed in MatLab using an open source quantum optics toolbox [36].

Figure 5 plots the calculated total absorbed power from the full master equation for three different values of \(|\varepsilon|^2\), which are expressed in units of \(\Gamma\), the modified spontaneous emission rate of the QD defined in Eq. (49). The semiclassical weak-field spectrum is also plotted. We use the identical parameters used to generate Fig. 4, and the dipole is set to be resonant with the MNP (\(\Delta_s = \Delta_c\)). Figure 5(a) plots the high cooperativity case where \(r = 10\) nm. As the input flux approaches the rate of one photon per modified lifetime of the QD \(|\varepsilon|^2 = \Gamma\) one can see that the reflection dip begins to decrease in contrast. The disappearance of the anti-resonance is caused by QD saturation. When the QD is saturated it can no longer interact with the MNP field, and the absorption spectrum approaches that of a bare MNP. We note, however, that even at weak driving fields of \(|\varepsilon|^2 = 0.001\Gamma\) the suppression of absorption is significantly worse than that predicted semiclassically.

A similar pattern is observed in the low cooperativity case shown in Fig. 5(b). Here, we set \(r = 25\) nm, resulting in a Fano line shape due to interference between driving field of the dipole and MNP. As the intensity of the driving field is increased, the Fano line shape deteriorates into the bare MNP absorption amplitude due to QD saturation. However, one can see that there is very poor agreement between the semiclassical and fully quantum calculations. The semiclassical Fano line shape produces a deeper antiresonance, while the quantum
line shape looks more like a resonant spike. Thus, the poor agreement between semiclassical and quantum calculations occurs both in the large and small cooperativity cases.

The inconsistency between the semiclassical and quantum theory is further illustrated in Fig. 6 where we set $r = 10$ nm and plot the minimum value of the antiresonance as a function of driving intensity. At high driving intensity the absorption approaches that of the bare MNP. As the driving field intensity decreases, the system makes a transition from the strong-field limit to the weak-field limit where the absorption is suppressed by an order of magnitude. As the driving intensity becomes very weak, the degree of suppression eventually becomes independent of pump power and achieves its steady weak-field value. However, this weak-field value is still more than an order of magnitude larger than the value predicted by semiclassical theory, as indicated by the dotted line.

To understand the cause of failure for the semiclassical theory, we plot the minimum value of the antiresonance for $r = 10$ nm as a function of the atomic cooperativity in Fig. 7. The solid line plots the absorbed power attained from the solution of the full master equation when both dephasing and spontaneous emission is present. The spontaneous emission rate is held fixed at $\gamma_s = 10^8$ s$^{-1}$ and the dephasing rate is modified. A second curve (dashed line) plots the absorbed power when there is only spontaneous emission. In this case $\gamma_s$ is adjusted to provide the same atomic cooperativities that were plotted when both dephasing and decay are present. According to semiclassical theory these two curves should be identical because the absorption rate depends only on the atomic cooperativity. The circles plot the absorbed power predicted by semiclassical theory. In all calculations the driving field was set to $|\epsilon|^2 = 10^{-6}$ to ensure we are in the weak-field limit so the calculation results can be properly compared to the analytical semiclassical weak-field expression.

The semiclassical theory agrees perfectly with the numerically calculated values when there is only spontaneous emission. The two curves lie on top of each other and are nearly indistinguishable. But when dephasing is incorporated into the calculations, the semiclassical theory severely underpredicts the amount of absorption.

![Figure 7](043845-9)
Figure 7 shows that the semiclassical mean-field theory approach is accurate for spontaneous emission but fails in the presence of dephasing. The problem occurs in the assumption that the field is quantified by a fixed amplitude $A$. In the weak-field limit with no dephasing the dipole and MNP are both linear scatterers, so if they are driven by a coherent field we have $\langle a \rangle = A$, where $A$ can now be interpreted as a coherent field amplitude. This result is correct even in the presence of spontaneous emission and MNP field damping due to the linear nature of the scattering. In this limit the semiclassical and quantum expressions will be equal. But if a dephasing process is present the cavity field is no longer a simple coherent field, it is an ensemble of coherent field amplitudes with different phases. In this case, averaging over phase jumps must be handled more carefully by averaging out the photon number operator, not the field amplitude.

V. RECONCILING THE SEMICLASSICAL AND QUANTUM THEORY IN THE WEAK-FIELD LIMIT

In this section we derive a corrected expression for the semiclassical weak-field absorption rate by applying the random phase jump method to the photon number operator instead of the field amplitude. We begin by writing the total absorbed power as $\Phi_{\text{abs}} = \Phi_{\text{scat}} + \Phi_{\text{dis}}$. For any practical MNP dipole system the power dissipated due to ohmic losses is significantly larger than the power dissipated due to scattering, because $\gamma_o \gg \gamma_s$ and $\gamma_o \gg \gamma_i$. Thus we can make the approximation $\Phi_{\text{abs}} \approx \Phi_{\text{dis}} \approx \kappa(a^\dagger a)$. The validity of this assumption will be confirmed by comparison to the full master equation calculations that take into account both scattering and ohmic losses.

We begin by taking the steady-state solution to Eq. (33) (we ignore the noise operators due to the cold reservoir assumption) which is given by

$$a = \frac{\sqrt{\kappa \epsilon - i g \sigma_-}}{i \Delta_c + \kappa/2}. \quad (63)$$

The average number of photons in the cavity is then given by

$$n = \langle a^\dagger a \rangle = \frac{4}{4\Delta_c^2 + \kappa^2} (\kappa \rho_{22} + 2g \kappa \epsilon \text{Im}[\rho_{21}] + \kappa \epsilon^2). \quad (64)$$

where $\text{Im}[]$ is the imaginary part of the bracketed expression. The above expression should be compared to the semiclassical expression given by

$$n_{\text{semi}} = |A|^2 = \frac{4}{4\Delta_c^2 + \kappa^2} (g^2 |\rho_{21}|^2 + 2g \sqrt{\kappa \epsilon} \text{Im}[\rho_{21}] + \kappa \epsilon^2). \quad (65)$$

We notice that the two expressions are nearly identical except that in the correct expression given in Eq. (64) the term $|\rho_{21}|^2$ is substituted by the term $\rho_{22}$, the population term of the excited state. When there is no dephasing we have the relation $|\rho_{21}|^2 = \rho_{11}\rho_{22}$, so in the weak field limit where $\rho_{11} \approx 1$ we have $|\rho_{21}|^2 = \rho_{22}$, and the two expressions are identical. Once dephasing is introduced, it reduces the off-diagonal term $\rho_{21}$ but does not affect the diagonal term $\rho_{22}$, and in this case the two expressions are not identical. We thus have a simple explanation for why the semiclassical theory is correct with only spontaneous emission but fails in the presence of dephasing.

To this point the expression in Eq. (64) is exact and does not make any weak-field approximation. However, we do not yet have a solution to this equation because $\rho_{21}$ is dependent on $n$ through the Maxwell-Bloch equations. But in the weak-field limit we can plug in the solutions given in Eqs. (57) and (58) into Eq. (64) to calculate $n$ and $\Phi_{\text{abs}}$. Figure 8 shows a comparison between the full master equation calculations and the corrected semiclassical equations using $\Phi_{\text{abs}} = \kappa n$, where $n$ is given by Eq. (64). Figure 8(a) plots the high atomic cooperativity case ($r = 10$ nm), whereas Fig. 8(b) plots the result for low atomic cooperativity ($r = 25$ nm). The driving intensity for the quantum calculations is set to $|\epsilon|^2 = 0.001 \Gamma$ to ensure that the solution is in the weak-field limit. The agreement between the quantum and corrected semiclassical calculations is nearly perfect. Since the full quantum calculations take into account both ohmic and scattering losses the strong agreement between the two calculations validates the assumption that scattering is negligibly small compared to ohmic losses when calculating the total absorbed power.

In Fig. 9 we repeat the calculations performed in Fig. 7, where we use the corrected semiclassical expression in order
defined as of central importance is the second-order correlation function addition to the absorption rate. For this purpose, a quantity investigate the quantum fluctuations of the MNP field in amplitudes we attain a weak-field expression that properly handles dephasing. by using the Heisenberg-Langevin equations directly and applying the random phase jump method to the cavity photon number expectation value instead of the field and dipole amplitudes we attain a weak-field expression that properly handles dephasing.

VI. QUANTUM FLUCTUATIONS

The full quantum optical formalism enables us to investigate the quantum fluctuations of the MNP field in addition to the absorption rate. For this purpose, a quantity of central importance is the second-order correlation function defined as

\[ g^{(2)}(\tau) = \frac{\langle \hat{a}(t)\hat{a}(t+\tau)\hat{a}(t+\tau)\hat{a}(t) \rangle}{\langle \hat{a} \rangle^2}. \] (66)

For a coherent state, \( g^{(2)}(\tau) = 1 \) for all times, indicating that each photon arrives independently. Fields with intensity noise (e.g., thermal states) will typically have a second-order correlation that exceeds 1 for small \( \tau \).

Figure 10 plots the second-order correlation of the MNP field for four different values of pumping intensity with \( d = 10 \) nm. The driving field is set to be at the minimum value of the antiresonance. Time is plotted in units of the modified spontaneous emission time \( \gamma^{-1} \). The second-order correlation function exhibits significant bunching on short time scales for the majority of curves. This bunching is due to the fact that if one photon is incident on the system, it will not excite the MNP due to the high atomic cooperativity. But if two photons arrive within the modified lifetime of the QD, the first photon will saturate the QD, increasing the probability that the second photon will excite the MNP. As the pumping intensity approaches \(|\epsilon|^2 = \Gamma\) the QD begins to saturate, which subsequently results in all photons being absorbed with nearly equal probability by the MNP. Thus, at higher intensities the autocorrelation approaches that of a bare MNP, which does not exhibit bunching.

The relationship between photon fluctuations and pumping intensity is seen more clearly in Fig. 11, which plots \( g^{(2)}(0) \) as a function of the pump rate normalized by \( \Gamma \). We plot this curve for three different values of the dephasing rate. In addition, a fourth curve plots the case where the dephasing rate is set to zero and the spontaneous emission rate is set to \( \gamma_s = 7.6 \) ns\(^{-1}\) to ensure a comparable atomic cooperativity. There is a significantly different bunching behavior at low powers when dephasing is present as opposed to the case with no dephasing. Without dephasing the bunching increases without bounds as the pump power is decreased. Introduction of dephasing results in the bunching leveling off at low pump powers and achieving a steady-state value. We note that this change in behavior is not caused by a difference in the atomic cooperativity, since the atomic cooperativity is the same for both cases.

![Figure 9](image9.png)

**FIG. 9.** (Color online) Minimum value of antiresonance for \( r = 10 \) nm as a function of atomic cooperativity. Solid line is full quantum calculation with dephasing and spontaneous emission. The dashed line is quantum calculation using only decay. Circles are corrected semiclassical calculations.

![Figure 10](image10.png)

**FIG. 10.** (Color online) Second-order correlation function for four different pumping intensity values.
VII. CONCLUSION

We have developed a cavity QED formalism for treating the interaction between a single MNP and a dipole emitter. We derived an analytical expression for the dipole-field coupling strength, the Purcell factor, and the atomic cooperativity, which are in good agreement with previous works that studied the radiative lifetime modification of dipoles in close proximity to MNPs of arbitrary shape [2,6–11]. Using the cQED formalism we derived a simple expression for the scattered and absorbed power of the MNP-dipole system when externally driven by a single-mode monochromatic field. In the weak-field limit we have shown that a large atomic cooperativity is possible, resulting in suppression of the scattering and absorption on resonance with the dipole. The strong-field limit was then investigated by numerical solution of the full master equation. At strong fields the interference effects are washed out by QD saturation and the absorption approaches that of a bare MNP. In addition, we found that proper handling of dephasing through the master equation approach results in significantly different behavior than semiclassical theory, even in the weak-field limit. This inconsistency was resolved by applying that random-phase-jump model to the full expectation value of the photon number operator, not just the field amplitude. This model was found to be in excellent agreement with the full quantum calculation in the weak-field limit.

The large cooperativities achievable in MNPs makes them an interesting candidate for studying cavity QED at the nanoscale. The formalism presented here provides a simple method to analyze such systems using a model that requires only a few parameters that can be either measured or calculated from first principles. This formalism can be extended in straightforward way to account for asymmetric nanoparticles such as nanorods and wires, enabling us to analyze a broad range of plasmonic devices interacting with atomic systems.

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APPENDIX A: RESPONSE OF MNP WITH DRUDE DISPERSION

When plugging Eq. (6) into Eq. (3) we attain the expression

$$E_{\text{MNP}} = \sum_{i} E_{0} G_{i}(r) \frac{\omega(\omega - i \gamma_{o}) \frac{\epsilon_{2}}{2 \epsilon_{2} + 1} + \omega_{0}^{2}}{\omega(\omega - i \gamma_{o}) - \omega_{0}^{2}},$$  \hspace{1cm} \text{(A1)}$$

where $\omega_{0} = \omega_{p}/\sqrt{2 \epsilon_{2} + 1}$. We consider the case where $\gamma_{o} \ll \omega$. In this limit the denominator of the above expression can be written as

$$\omega(\omega - i \gamma_{o}) - \omega_{0}^{2} \approx 2 \omega[(\omega - \omega_{0}) - i \gamma_{o}/2].$$  \hspace{1cm} \text{(A2)}$$

which is the standard approximation for a high-$Q$ resonator. In this limit the electric field response will be very small unless $\omega \approx \omega_{0}$. Using the assumption that $\omega \approx \omega_{0} \gg \gamma_{o}$ the numerator in Eq. (A1) can be simplified to

$$\omega(\omega - i \gamma_{o}) \frac{\epsilon_{2}}{2 \epsilon_{2} + 1} + \omega_{0}^{2} \approx \omega_{0}^{2} \frac{3 \epsilon_{1}}{2 \epsilon_{2} + 1}. $$  \hspace{1cm} \text{(A3)}$$

Plugging these approximations for the numerator and denominator back into Eq. (A1) we attain Eq. (7).

APPENDIX B: DERIVATION OF HEISENBERG-LANGEVIN EQUATIONS OF MOTION

The Heisenberg equations of motion for the field and dipole operators can be calculated directly from Eq. (31) and are given by

$$\frac{d\hat{a}}{dt} = -(i \omega_{0} + \kappa/2)\hat{a} - i \gamma_{a} - \sqrt{\kappa} \hat{e} e^{i \omega t}$$

$$- T_{1} \int d\omega \hat{b}_{\omega} - T_{2} \int d\omega \hat{c}_{\omega},$$  \hspace{1cm} \text{(B1)}$$

$$\frac{d\sigma_{e}}{dt} = -(i \Delta_{d} + \gamma_{s}/2)\sigma_{-} + i \gamma_{a} \sigma_{\lambda} - \Omega \sigma_{e} e^{i \omega t} - T_{3} \int d\omega \hat{b}_{\omega},$$  \hspace{1cm} \text{(B2)}$$

$$\frac{d\sigma_{\lambda}}{dt} = 2 i g(\sigma_{-} - \sigma_{+}, \hat{a}) + 2 \Omega(\sigma_{-} + \sigma_{+})$$

$$- 2 T_{3} \int d\omega \hat{b}_{\omega}^{\dagger} \sigma_{-} + \sigma_{-} \hat{b}_{\omega},$$  \hspace{1cm} \text{(B3)}$$

$$\frac{d\hat{b}_{\omega}}{dt} = -i \omega \hat{b}_{\omega} + T_{1} \hat{a} + T_{2} \sigma_{-},$$  \hspace{1cm} \text{(B4)}$$

$$\frac{d\hat{c}_{\omega}}{dt} = -i \omega \hat{c}_{\omega} + T_{2} \hat{a}.$$  \hspace{1cm} \text{(B5)}$$

Equations (B4) and (B5) can be directly integrated to give

$$\hat{b}_{\omega}(t) = \hat{b}_{\omega}(0) e^{-i \omega t} + \int_{0}^{t} (T_{1} \hat{a} + T_{3} \sigma_{-}) e^{-i \omega(t-t')} dt'$$  \hspace{1cm} \text{(B6)}$$

$$\hat{c}_{\omega}(t) = \hat{c}_{\omega}(0) e^{-i \omega t} + \int_{0}^{t} T_{2} \hat{a} e^{-i \omega(t-t')} dt'.$$  \hspace{1cm} \text{(B7)}$$

Plugging the above solutions into Eqs. (B1) and (B2) we obtain

$$\frac{d\hat{a}}{dt} = -(i \omega_{0} + \kappa/2)\hat{a} - i \gamma_{a} - \sqrt{\kappa} \hat{e} e^{i \omega t} - \pi (T_{1}^{2} + T_{2}^{2}) \hat{a}$$

$$- \pi T_{1} T_{3} \sigma_{-} - \int d\omega (T_{1} \hat{b}_{\omega}(0) + T_{2} \hat{c}_{\omega}(0)) e^{-i \omega t},$$  \hspace{1cm} \text{(B8)}$$

$$\frac{d\sigma_{e}}{dt} = -(i \Delta_{d} + \gamma_{s}/2)\sigma_{-} + i \gamma_{a} \sigma_{\lambda} - \Omega \sigma_{e} e^{i \omega t} - \pi T_{2} \sigma_{-}$$

$$- \pi T_{1} T_{3} \sigma_{-} - \int d\omega \hat{b}_{\omega}(0) e^{-i \omega t},$$  \hspace{1cm} \text{(B9)}$$

$$\frac{d\sigma_{\lambda}}{dt} = 2 i g(\sigma_{-} - \sigma_{+}, \hat{a}) + 2 \Omega(\sigma_{-} + \sigma_{+}) - 2 \pi T_{3} \sigma_{-} + 1$$

$$- 2 \pi T_{1} T_{3} (\hat{a} \sigma_{-} + \sigma_{+} a) - T_{3} \int d\omega \hat{b}_{\omega}(0) \sigma_{-} e^{i \omega t} + \sigma_{-} \hat{b}_{\omega}(0) e^{-i \omega t}. $$  \hspace{1cm} \text{(B10)}$$

We define $\gamma_{r} = 2 \pi T_{1}^{2}$, $\gamma_{o} = 2 \pi T_{2}^{2}$, and $\gamma_{s} = 2 \pi T_{3}$. In addition we define the noise operators

$$\hat{f} = - \int d\omega (T_{1} \hat{b}_{\omega}(0) + T_{2} \hat{c}_{\omega}(0)) e^{i(\omega - \omega')t},$$  \hspace{1cm} \text{(B11)}$$

$$\hat{g} = -T_{3} \int d\omega \hat{b}_{\omega}(0) e^{i(\omega - \omega')t}$$  \hspace{1cm} \text{(B12)}$$

$$\hat{h} = -2 T_{3} \int d\omega \hat{b}_{\omega}(0) \sigma_{-} e^{i \omega t} + \sigma_{-} \hat{b}_{\omega}(0) e^{-i \omega t}.$$  \hspace{1cm} \text{(B13)}$$
Using these definitions, and making the transformation into the frame rotating at the driving frequency $\omega$ in Eqs. (B8) and (B9), we obtain the Heisenberg equations of motion given in Eqs. (33)–(35).

APPENDIX C: RADIATIVE DECAY OF THE MNP

If a dipole is excited by an incident electric field of the form $E_0(r,t) = \Delta \delta(t) \hat{z}$ the electric field at time $t > 0$ is given by

$$E(r) = \alpha \sin \omega_0 t \begin{cases} z \left( \frac{r}{R} \right) & r < R \\ \left( 2 \cos \theta \hat{r} + \sin \theta \hat{\theta} \right) & r > R \end{cases}$$

where $\alpha = \lambda \eta \omega_0 / 2$. The energy stored in the electric field (averaged over an optical cycle) is given by

$$\langle W \rangle = \frac{\epsilon_0 \alpha^2}{4} \int d^3r \frac{\epsilon_0}{2} |G(r)|^2 \text{Re} \left( \frac{d}{d\omega} (\omega) \right)$$

$$= \frac{\epsilon_0 \alpha^2}{2} V_m (\epsilon_2 + 1).$$

Outside the MNP, the electric field takes on the form of a dipole field produced by a dipole moment $p = 4\pi \epsilon_0 \epsilon_2 R^3 \alpha$ oscillating at the frequency $\omega_0$. We can express the energy in the field in terms of this dipole moment as

$$\langle W \rangle = \frac{p^2 (\epsilon_2 + 1)}{24\pi \epsilon_0 \epsilon_2^2 R^4}.$$  \hfill (C1)

In the limit that the MNP is much smaller than the wavelength of light we can treat it as a point dipole. The amount of power radiated by this point dipole is given by the Larmor formula:

$$\frac{d(W)}{dt} = -\frac{\mu_0 \omega_0 p^2}{12\pi c}.$$ \hfill (C2)

The above two equations can be written as

$$\frac{d(W)}{dt} = -\gamma_r \langle W \rangle,$$ \hfill (C3)

where

$$\gamma_r = \frac{2\omega_0^2 \epsilon_2^2 R_0^3}{c^3 (2\epsilon_2 + 1)}.$$ \hfill (C4)

APPENDIX D: DERIVATION OF OUTPUT FLUX OPERATOR

Plugging Eq. (B6) into Eq. (36) we obtain

$$\hat{b}_{\text{out}}(t) = \frac{1}{\sqrt{2\pi}} \int d\omega \hat{b}_\omega(0)e^{-i\omega t}$$

$$+ \frac{1}{\sqrt{2\pi}} \int d\omega \int_{t'}^t d\tau (T_1 \hat{a} + T_3 \sigma_-)e^{-i\omega(t-\tau)}$$

$$= \frac{1}{\sqrt{2\pi}} \int d\omega \hat{b}_\omega(0)e^{-i\omega t} + \sqrt{2\pi}(T_1 \hat{a} + T_3 \sigma_-).$$

Because the output modes are initially assumed to be in vacuum, the terms proportional to $\hat{b}_\omega(0)$ can be disregarded as they will always vanish when operating on the initial vacuum state. Using the fact that $\gamma_r = 2\pi T_1^2$ and $\gamma_s = 2\pi T_3^2$ we directly obtain the expression in Eq. (37).