Communication

# Dynamics of Nano-Particles Inside an Optical Cavity in the Quantum Regime 

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#### Abstract

We investigate the optomechanical effect on a single nano-particle inside an optical cavity, by deriving the optical forces acting on the nano-particle by the cavity from quantum theory. We obtain the steady state of the system and found that the force contains three terms associated with the gradient force, the back-action force resulting from the intra-cavity photon energy change, as well as the reactive force associated with the coupling between the external field and the cavity. Moreover, we solve the dynamical system for a dielectric particle in a small mode volume cavity, which is characterized by a quasi-periodic pattern. These results are important for understanding the control of various types of levitated nano-particles through optomechanical coupling.


Keywords: optical trapping; optical forces; quantum optomechanics; cavity quantum electrodynamics; dynamical evolution

## 1. Introduction

Since the first demonstration that light can trap and manipulate small particles [1,2], extensive research has been continued in the field [3-9]. The conventional optical tweezer trapping is, however, bounded by a diffraction limit. To overcome this limitation [10], different devices have been proposed and experimentally tested to carry out the optical trapping; see, e.g., optical microcavities [11-13], plasmonic/metallic structures [14-16], and photonic crystal structures for multiple particle trapping [17,18]. Recently, particular attention has been paid to levitated particles of optomechanical coupling, because the mechanical quality can be significantly improved to suppress the decoherence from the thermal noise effect of the environment [19-22]. In addition to the nano-particles of simple dielectric materials, current research has been forwarded to those of internal degrees of freedom such as optically active defects [23-26], to see the novel phenomena related to their properties. Among various research directions, the biggest progress has been made in the realization of the ground quantum state of trapped particles [27,28]. However, except for a few studies about the linearized dynamics [29-32], almost no research has been undertaken to understand the nonlinear dynamical behaviors of trapped particles. In this work, we present a complete calculation of the potential for trapping the particle based on full quantum theory and study the nonlinear dynamics of a trapped nano-particle in terms of the derived potential. We apply this formalism to a cavity with small mode volume and a low quality factor and find that, depending on the initial condition of the particle, the resulting dynamical pattern can be periodic and quasi-periodic. This kind of dynamics can be explored experimentally by using photonic crystal cavities [33].

## 2. Optical Force Calculation in the Quantum Regime

Let us assume a cavity with the resonant frequency $\omega_{c 0}$ and optical mode $E_{c}(r)$. When we introduce a particle at the position $x_{0}$, it can be seen as a perturbation to the dielectric
constant of the cavity. This perturbation depends on the particle position and the particle polarizability $\operatorname{Re}(\alpha(\omega))$, which in general is frequency-dependent. Then, the cavity will have a new cavity resonance $\tilde{\omega}_{c}\left(x_{0}\right)$ depending on the particle position.

The Hamiltonian of the system can be written as a sum of the kinetic energy, the cavity energy, the coupling with the external source, and the polarization energy of the particle [34-36]. In a rotating frame at the pump frequency $\omega$, the Hamiltonian of the system reads as:

$$
\begin{array}{r}
H=\frac{p^{2}}{2 m}+\hbar\left(\tilde{\omega}_{c}\left(x_{0}\right)-\omega\right) a^{\dagger} a+i \hbar \int \gamma\left(\omega, \tilde{\omega}_{c}\right)\left(a^{\dagger} s-a s^{\dagger}\right) d \omega \\
-\frac{1}{2} \int_{V_{p}} P(y) E_{c}(y) d y \tag{1}
\end{array}
$$

where $a / a^{\dagger}$ is the operator creating/annihilating a cavity photon, and $s / s^{\dagger}$ represents the pumping source. Moreover, $\gamma\left(\omega, \tilde{\omega}_{c}\right)$ is a coupling constant, and $P$ is the polarization of the particles, while the last integral is over the particle occupation space $V_{p}$. For a monochromatic source, the coupling to the external source (third term in Equation (1)) can be expressed as

$$
\begin{equation*}
i \hbar \int \gamma\left(\omega, \tilde{\omega}_{c}\right)\left(a^{\dagger} s-a s^{\dagger}\right) d \omega=i \hbar \sqrt{2 \gamma_{e}(x)} s_{i n}\left(a^{\dagger}-a\right) \tag{2}
\end{equation*}
$$

where $\gamma_{e}$ is the cavity decay rate, so that $s_{i n}=\sqrt{\frac{P_{i n}}{\hbar \omega}}$ is the photon flux from the source of the power $P_{i n}$. In the last term, we assume that the particle has a polarizability $P=\operatorname{Re}\left(\alpha\left(\tilde{\omega}_{c}\right)\right) E_{c}$. Then, the interaction Hamiltonian is

$$
\begin{align*}
H_{\text {int }}=-\frac{1}{2} \int_{V_{p}(x)} P(y) E_{c}(y) d y=- & \frac{\operatorname{Re}\left(\alpha\left(\tilde{\omega}_{c}\right)\right)}{2} \int_{V_{p}(x)}\left|E_{c}(y)\right|^{2} d y  \tag{3}\\
& =-\hbar \tilde{\omega}_{c} \operatorname{Re}\left(\alpha\left(\tilde{\omega}_{c}\right)\right) f\left(x_{0}\right) a^{\dagger} a
\end{align*}
$$

where $f\left(x_{0}\right)$ is a function that depends on the position $x_{0}$ of the particle and is determined by the integral of the field over the particle volume.

To calculate the optomechanical force at the position $x_{0}$, it is necessary to know the change in energy for a small displacement $x$ from $x_{0}$. The interaction Hamiltonian can be written as

$$
\begin{align*}
H_{\text {int }}(x)=-\hbar \tilde{\omega}_{c} \operatorname{Re}\left(\alpha\left(x_{0}\right)\right) f\left(x_{0}\right) a^{\dagger} a-x \hbar \tilde{\omega}_{c}[ & \operatorname{Re}\left(\alpha\left(x_{0}\right)\right) \frac{\partial f(x)}{\partial x} \\
+ & \left.f\left(x_{0}\right) \frac{\partial \operatorname{Re}(\alpha(x))}{\partial x}\right] a^{\dagger} a \tag{4}
\end{align*}
$$

where, by using the complex polarizability $\alpha=\alpha_{0} /\left[1-i k^{3} \alpha_{0} /\left(6 \pi \epsilon_{0} \epsilon_{m}\right)\right]$, the time-averaged total gradient, absorbing and scattering force in the nano-particle can be taken into account [37]. It represents the average energy on a time-harmonic electromagnetic field by a small particle.

### 2.1. Low Polarizability

At low polarizability, the detuning is small, so the Hamiltonian can be written as (see Appendix A for details):

$$
\begin{equation*}
H(x)=H_{0}-x \hbar \omega_{0} \frac{\partial[\operatorname{Re}(\alpha) f(x)]}{\partial x} a^{\dagger} a \tag{5}
\end{equation*}
$$

where $H_{0}$ is the energy of the system for a particle at $x=x_{0}$ and $\omega_{0}=\tilde{\omega}_{c}\left(x_{0}\right)$. We have assumed that, at a fixed frequency, the energy in the cavity is constant around a given $x_{0}$
and, therefore, the coupling coefficient is also a constant. Solving the quantum Langevin equations $[38,39]$ with the Hamiltonian given in Equation (5), we obtain the following:

$$
\begin{gather*}
\frac{\partial a}{\partial t}=i\left[\omega-\omega_{0}+\left(\omega_{0} \frac{\partial[\operatorname{Re}(\alpha(x)) f(x)]}{\partial x}\right) x\right] a-\left(\gamma_{i}+\gamma_{e}\right) a+\sqrt{2 \gamma_{e}} s_{i n}  \tag{6}\\
\frac{\partial p}{\partial t}=\hbar \omega_{0}\left(\frac{\partial[\operatorname{Re}(\alpha) f(x)]}{\partial x}\right) a^{+} a, \tag{7}
\end{gather*}
$$

with $\gamma_{i}$ being the particle intrinsic loss. We have defined $\omega_{c}=\omega_{0}(1-\operatorname{Re}(\alpha(x)) f(x))$ as the additional detuning. This extra detuning, $\omega_{c}$, can also be derived from the HellmannFeynman theorem, as presented in the open cavity analysis [10].

The steady state is obtained with $d a / d t=0$, so that

$$
\begin{gather*}
a_{s}\left(\omega, x_{0}\right)=\frac{\sqrt{2 \gamma_{e}} s_{i n}}{\left(\gamma_{e}+\gamma_{i}\right)-i\left(\omega-\omega_{0}\right)}  \tag{8}\\
F_{s}\left(x_{0}\right)=\left.\hbar \omega_{0} \frac{\partial[\operatorname{Re}(\alpha(x)) f(x)]}{\partial x}\right|_{x=x_{0}}\left|a_{s}\right|^{2} \tag{9}
\end{gather*}
$$

are the steady-state values. In the case of dielectric particles, there are $\gamma_{i}=0, \frac{\partial(\operatorname{Re}(\alpha))}{\partial x}=0$ and $Q=\frac{\omega_{0}}{2 \gamma_{e}}$. The force expression (Equation (9)) is then reduced to:

$$
\begin{equation*}
F_{s}\left(x_{0}\right)=\left.\frac{2 Q \omega_{0}^{2} P_{\text {in }} \operatorname{Re}(\alpha)}{\omega\left[\omega_{0}^{2}+4 Q^{2}\left(\omega-\omega_{0}\right)^{2}\right]} \frac{\partial\left[\left|E_{0}(x)\right|^{2}\right]}{\partial x}\right|_{x=x_{0}} \tag{10}
\end{equation*}
$$

where $P_{\text {in }}$ is the source power. This expression is similar to the one postulated using open cavity analysis [10], with a coupling coefficient

$$
\begin{equation*}
T(\omega)=\frac{\omega_{0}^{2}}{\omega_{0}^{2}+4 Q^{2}\left(\omega-\omega_{0}\right)^{2}} \tag{11}
\end{equation*}
$$

### 2.2. High Polarizability

In order to calculate the optomechanical force at the position $x_{0}$ for particles with high polarizability or frequency-dependent polarizability, we can again estimate the change in energy for a small displacement $x$ around $x_{0}$. If the cavity is at frequency $\omega_{0}$, the additional cavity detuning and change in the cavity-coupling coefficient will produce a change in the photon field energy coupled to the cavity, producing the additional force. We can estimate the change in cavity frequency and coupling by using the following Taylor expansions around $x_{0}$ :

$$
\begin{gather*}
\omega_{c}(x)=\omega_{0}+\left.x \frac{\partial \omega_{c}}{\partial x}\right|_{x=x_{0}}  \tag{12}\\
\sqrt{2 \gamma_{e}\left(\omega_{c}(x)\right)}=\left(\sqrt{2 \gamma_{e}\left(\omega_{0}\right)}+\frac{x}{\sqrt{2 \gamma_{e}\left(\omega_{c}\right)}} \frac{\partial \gamma_{e}}{\partial x}\right) . \tag{13}
\end{gather*}
$$

For the photon number in the cavity, assuming the electromagnetic field profile is unperturbed, we can calculate the change by using the canonical quantization of the field in the form [40-42]:

$$
\begin{equation*}
a(\omega)=\frac{1}{\sqrt{2 \hbar \omega}}\left(\omega X_{c}+i P_{c}\right) \tag{14}
\end{equation*}
$$

for canonical operators $X_{c}$ and $P_{c}$. Thus, the expansion of the operator around $x_{0}$ is

$$
\begin{equation*}
a(x)=a\left(x_{0}\right)+x \frac{\partial a}{\partial \omega_{c}} \frac{\partial \omega_{c}}{\partial x}=a\left(x_{0}\right)+\left.\frac{x}{2 \omega_{c}} a^{\dagger} \frac{\partial \omega_{c}}{\partial x}\right|_{x=x_{0}} \tag{15}
\end{equation*}
$$

After incorporating all the terms into the Hamiltonian, together with applying the rotation wave approximation and neglecting terms involving two photon processes (see Appendix A), we obtain the Hamiltonian and the quantum Langevin equation:

$$
\begin{gather*}
H(x)=H_{0}+x\left[\hbar \frac{\partial \omega_{c}}{\partial x} a^{\dagger} a-i \hbar \frac{\sqrt{2 \gamma_{e}}}{2 \omega_{c}}\left(a^{\dagger}-a\right) s_{i n} \frac{\partial \omega_{c}}{\partial x}+\frac{i \hbar}{\sqrt{2 \gamma_{e}}}\left(a^{\dagger}-a\right) s_{i n} \frac{\partial \gamma_{e}}{\partial x}\right] \\
+x\left[-\hbar \omega_{c} \operatorname{Re}(\alpha) \frac{\partial f(x)}{\partial x} a^{\dagger} a-\hbar \omega_{c} f\left(x_{0}\right) \frac{\partial \operatorname{Re}(\alpha(x))}{\partial x} a^{\dagger} a-\hbar \omega_{c} f\left(x_{0}\right) \operatorname{Re}\left(\alpha\left(x_{0}\right)\right) \frac{\partial \omega_{c}}{\partial x} a^{\dagger} a\right],  \tag{16}\\
\frac{\partial p}{\partial t}=\hbar \omega_{c}\left[\frac{\partial[\operatorname{Re}(\alpha) f(x)]}{\partial x}+\frac{f\left(x_{0}\right) \operatorname{Re}\left(\alpha\left(x_{0}\right)\right)}{\omega_{c}} \frac{\partial \omega_{c}}{\partial x}\right] a^{\dagger} a-\hbar \frac{\partial \omega_{c}}{\partial x} a^{\dagger} a \\
+i \hbar \frac{\sqrt{2 \gamma_{e}}}{2 \omega_{c}}\left(a^{\dagger}-a\right) \sin _{i n} \frac{\partial \omega_{c}}{\partial x}-\frac{i \hbar}{\sqrt{2 \gamma_{e}}}\left(a^{\dagger}-a\right) s_{i n} \frac{\partial \gamma_{e}}{\partial x} . \tag{17}
\end{gather*}
$$

Similarly to the low polarizability case, we obtain the steady-state solutions:

$$
\begin{align*}
& a_{s}\left(\omega, x_{0}\right)=\frac{\sqrt{2 \gamma_{e}} s_{i n}}{\left(\gamma_{e}+\gamma_{i}\right)-i\left(\omega-\omega_{c}\right)}  \tag{18}\\
F_{s}\left(x_{0}\right)=\quad & \hbar \omega_{c}\left(\frac{\partial[\operatorname{Re}(\alpha(x)) f(x)]}{\partial x}\right)\left|a_{s}\right|^{2} \\
& +\hbar\left[f\left(x_{0}\right) \operatorname{Re}\left(\alpha\left(x_{0}\right)\right)\left|a_{s}\right|^{2}+i \frac{\sqrt{2 \gamma_{e}}}{2 \omega_{c}}\left(a^{\dagger}-a\right) s_{i n}-\left|a_{s}\right|^{2}\right] \frac{\partial \omega_{c}}{\partial x} \\
& -\frac{i \hbar}{\sqrt{2 \gamma_{e}}}\left(a_{s}^{\dagger}-a_{s}\right) s_{i n} \frac{\partial \gamma_{e}}{\partial x} . \tag{19}
\end{align*}
$$

The first term in Equation (19) is associated with the gradient force, and the second is the back-action resulting from the intra-cavity photon energy change, and the last term is the reactive force from the coupling between the external field and the cavity. Finally, the force at the position $x_{0}$ can be expressed as

$$
\begin{align*}
& F_{s}\left(x_{0}\right)=-\left.\frac{2 \gamma_{e} P_{i n}}{\omega\left[\left(\gamma_{e}+\gamma_{i}\right)^{2}+\left(\omega-\omega_{c}\right)^{2}\right]} \frac{\partial \omega_{c}}{\partial x}\right|_{x=x_{0}} \\
&+\left.\frac{\gamma_{e} P_{i n} \operatorname{Re}\left(\alpha\left(x_{0}\right)\right)\left|E_{0}(x)\right|^{2}}{\omega\left[\left(\gamma_{e}+\gamma_{i}\right)^{2}+\left(\omega-\omega_{c}\right)^{2}\right]} \frac{\partial \omega_{c}}{\partial x}\right|_{x=x_{0}} \\
&-\left.\frac{2\left(\omega-\omega_{c}\right) P_{i n}}{\omega\left[\left(\gamma_{e}+\gamma_{i}\right)^{2}+\left(\omega-\omega_{c}\right)^{2}\right]} \frac{\partial \gamma_{e}}{\partial x}\right|_{x=x_{0}} \\
&+\left.\frac{\omega_{c} \gamma_{e} P_{i n}}{\omega\left[\left(\gamma_{e}+\gamma_{i}\right)^{2}+\left(\omega-\omega_{c}\right)^{2}\right]} \frac{\partial\left[\operatorname{Re}(\alpha(x))\left|E_{0}(x)\right|^{2}\right]}{\partial x}\right|_{x=x_{0}} . \tag{20}
\end{align*}
$$

## 3. Dynamics of a Dielectric Particle in the Optical Cavity

To study the dynamics of the particle, we look at the simplest case of a dielectric particle, $\gamma_{i}=0$ and $Q=\frac{\omega_{0}}{2 \gamma_{e}}$, to simplify the force expression Equation (20) to Equation (10). In addition, we consider a simple cavity of length $L=\lambda / 2$, with $\lambda$ being the pumping wavelength, to have a Gaussian distribution of the field in the lateral directions, with
$w_{c}=\lambda / 2 N A$ for $N A=0.7$ and a quality factor $Q=2000$. This exemplary cavity demonstrates how the model works for a generic category of the cavities with small mode volume and a low quality factor. Since the radius $r$ of a spherical particle can be much smaller than the parameter $\left(w_{c}\right)$, one can approximate the electromagnetic field profile as

$$
\begin{equation*}
\left|E_{0}(x, y)\right|^{2}=\frac{1}{E_{V}} \exp \left(-\frac{2 y^{2}}{w_{c}^{2}}\right) \cos ^{2}\left(\frac{\pi x}{L}\right) \tag{21}
\end{equation*}
$$

where $E_{V}$ represents the total energy in the cavity. It should be noted that the cavity detuning, as a function of particle position inside the cavity, is given in [10] (see Appendix B for details):

$$
\begin{equation*}
\omega_{c}(x)-\omega=-\frac{\omega_{c}(x)|E(x)|^{2} \operatorname{Re}(\alpha)}{\int_{V c} \epsilon_{c}|E(x)|^{2} d V} \tag{22}
\end{equation*}
$$

We also consider a pumping source with a wavelength $\lambda=1.55 \mu \mathrm{~m}$ and a power $P_{\text {in }}=5 \mathrm{~mW}$. The particle has $r=20 \mathrm{~nm}$, dielectric permittivity $\epsilon_{p}=12$ and density $2.33 \mathrm{~g} / \mathrm{cm}^{3}$.

Figure 1 shows the optical force and associated potential as a function of the particle position. The force with a nonlinear characteristic shows a symmetric landscape, making us to go beyond the linear regime.


Figure 1. (a) Force x-component $\left(F_{x}\right)$. (b) Force y-component $\left(F_{y}\right)$. (c) Potential energy of the particle in the cavity. Black color lines represent equipotential lines. Horizontal axis in units of $L / 2$. Vertical axis in units of $w_{c}$. Force in pN . Particle potential energy in units of aJ.

For simplicity, we can reduce the phase space of the system to the section $\left(x, p_{x}, y, p_{y}\right)$ by fixing $\left(z, p_{z}\right)=(0,0)$; the extension to the whole space of six dimensions $\left(x, p_{x}, y, p_{y}, z, p_{z}\right)$ is straightforward. Let us consider the particle with small drag and gravitation force; otherwise, in a damped system, the rich dynamics will be lost as the particle stabilizes only in a terminal velocity. In general, the dynamics of the system manifests by the solution $\left(x, p_{x}, y, p_{y}\right)$ to the following differential equations:

$$
\begin{array}{r}
\dot{\vec{x}}=\frac{\vec{p}}{m} \\
\dot{\vec{p}}=\hbar \omega_{c}\left|a_{s}(x)\right|^{2} \nabla[\operatorname{Re}(\alpha(x)) f(x)] . \tag{24}
\end{array}
$$

The initial condition $\left(x_{0}, p_{x, 0}, y_{0}, p_{y, 0}\right)=\left(x_{\text {ini }}, 0,0,0\right)$ or $\left(x_{0}, p_{x, 0}, y_{0}, p_{y, 0}\right)=\left(0,0, y_{\text {ini }}, 0\right)$, where $0<x_{i n i}<L / 2$ and $0<y_{i n i}<w_{c}$ given the force/potential's symmetry (see Figure 1), will lead to different trajectories of the particle. From a given initial conditions ( $x_{0}, p_{x 0}, y_{0}, p_{y 0}$ ), the dynamical system is solved numerically to obtain the behavior of the levitated particle. For the initial conditions $\left(x_{0}, 0, y_{0}, 0\right)$ with $x_{0} \ll L / 2$ and $y_{0} \ll w_{c}$, the particle shows
periodic trajectories as expected from the approximate linear potential at a short displacement. Given the small mode volume of the cavity, the particle will experience the full potential, and then, we should go beyond the regime of periodic motion.

The particle will display the periodic oscillations of complicated patterns if the particle is confined in one dimension such as along the X -axis, Y -axis. However, when its initial is more general with the position $\left(x_{0}, 0, y_{0}, 0\right)$ in the cavity, the realized trajectories will much more complicated as the quasi-periodic ones as shown with the Poincare section $\left(x_{0}, p_{x, 0}, y_{0}, p_{y, 0}\right)=\left(x_{i n i}, 0, w_{c}, 0\right)$ in Figure 2a. The displayed Poincare sections are totally symmetric with respect to both position and momentum of the particle motion. Moreover, the change of the initial condition modifies its profile while maintaining its symmetry, indicating that the quasi-periodic characteristic is typical to this system. In order to get the further information about the motion, we calculate the power spectral density (PSD) of the moving particle along each direction. This efficient technique allows one to explore the different regimes such as periodic, quasi-periodic, and chaotic in complex systems [43]. In a special situation of pure periodic motion, the PSD will have the form of a single peak. However, in Figure 2b, the PSD displays a spectrum of multiple peaks even for one particular case of $\left(x_{i n i}, 0, w_{c}, 0\right)$. The demonstrated multi-peak pattern indicates the multiple frequency components in oscillation, which are typical for a quasi-periodic motion. Such PSD from the projection of a two dimensional motion implies rather complex quasiperiodic trajectories by nature. These trajectories can be well adjusted by tuning the system parameters.


Figure 2. (a) Poincare section of the $P y$ of the particle at $\left(x_{0}, p_{x 0}, y_{0}, p_{y 0}\right)=\left(x_{i n i}, 0, w_{c}, 0\right)$ with $x_{\text {ini }}=\frac{L}{2}\left(\frac{i}{6}\right), i=1, \ldots, 5$. The momentum is in units of $\mathrm{fg} / \mathrm{s}$ and y in units of $w_{c}$. (b) Power spectral density spectrum for the $y$ component of the position of the particle for $x_{i n i}=\frac{L}{2}\left(\frac{5}{6}\right)$.

## 4. Discussion

The concerned optomechanical system, consisting of a levitating nano-particle in an optical cavity with a small mode volume, displays periodic and quasi-periodic motions depending on its initial conditions. In the system we present as an example, the small detuning is driven by the particle position but, for the systems where the detuning is higher [13,44], we need to consider all the terms in Equation (20) rather than the last term only, i.e., including the back-action and reactive forces. The complete dynamical picture was not considered before.

Beyond the scenario of the steady state, the dynamical behaviour of the levitated particle could be rather complicated due to the nonlinearity in the interaction potential.Through the stronger interplay between the cavity field and mechanical motion, the adjusted lasercavity detuning and optical coupling can lead to a chaotic motion [45,46]. In general, the equations of motion in Equation (6) are nonlinear, and the variables of the equations of motion are greater than three if we put them in a standard form of an autonomous system.

In the extreme situation, chaos would manifest for certain detuning and laser power, as seen in other optomechanical systems [46-48].

## 5. Conclusions

We have investigated mechanical forces acting on a single nano-particle inside an optical cavity. We derive the optical forces acting on the nano-particle from a full quantum theory. The force contains three terms associated with the gradient force, the back-action force resulting from the intra-cavity photon energy change, as well as the reactive force related to the coupling between the external field and the cavity. The dynamical system of the dielectric particle is solved by considering these forces in a small mode volume cavity. Particularly, a quasi-periodic behavior of the particle displays a symmetric Poincare map and a multi-peak power density spectrum of the displacement. Such rich dynamical behaviors could be further explored in the systems where the back-action force and reactive force are larger compared to the gradient force, which can be realized for different types of levitated resonant particles, high-Q cavities, and more complex cavity field profiles.

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## Appendix A

In this section, all the derivatives are evaluated at $x=x_{0}$ (so, they are numbers, no operators). Omitting second-order terms ( $x^{2}$, aa and $a^{\dagger} a^{\dagger}$ ), we Taylor expand each term of the Hamiltonian (Equation (1)) using Equations (12)-(14) to take into account the extra cavity detuning:

$$
\begin{equation*}
\hbar\left(\omega_{c}(x)-\omega\right) a^{\dagger}(x) a(x)=\hbar\left(\omega_{c}\left(x_{0}\right)-\omega+x \frac{\partial \omega_{c}}{\partial x}\right) a^{\dagger} a \tag{A1}
\end{equation*}
$$

similarly,

$$
\begin{array}{r}
\sqrt{2 \gamma_{e}(x)} s_{i n}\left(a^{\dagger}(x)-a(x)\right)=\left(\sqrt{2 \gamma_{e}}\left(a^{\dagger}-a\right)+\frac{x}{\sqrt{2 \gamma_{e}}} \frac{\partial \gamma_{e}}{\partial x}\left(a^{\dagger}-a\right)\right. \\
\left.-\frac{x \sqrt{2 \gamma_{e}}}{2 \omega_{c}} \frac{\partial \omega_{c}}{\partial x}\left(a^{\dagger}-a\right)\right) s_{i n} \tag{A2}
\end{array}
$$

and the last term:

$$
\begin{array}{r}
-\hbar \omega_{c}(x) \operatorname{Re}(\alpha(x)) f(x) a^{\dagger} a=-\hbar\left[\omega_{c} \operatorname{Re}(\alpha) f\left(x_{0}\right)\right. \\
\left.+x \omega_{c} \frac{\partial \operatorname{Re}(\alpha) f\left(x_{0}\right)}{\partial x}+x \operatorname{Re}(\alpha) f\left(x_{0}\right) \frac{\partial \omega_{c}}{\partial x}\right] \tag{A3}
\end{array}
$$

Therefore, the Hamiltonian can be written as Equation (16):

$$
\begin{array}{r}
H(x)=H_{0}+x\left[\hbar \frac{\partial \omega_{c}}{\partial x} a^{\dagger} a-i \hbar \frac{\sqrt{2 \gamma_{e}}}{2 \omega_{c}}\left(a^{\dagger}-a\right) s_{i n} \frac{\partial \omega_{c}}{\partial x}+\frac{i \hbar}{\sqrt{2 \gamma_{e}}}\left(a^{\dagger}-a\right) s_{i n} \frac{\partial \gamma_{e}}{\partial x}\right] \\
+x\left[-\hbar \omega_{c} \operatorname{Re}(\alpha) \frac{\partial f(x)}{\partial x} a^{\dagger} a-\hbar \omega_{c} f\left(x_{0}\right) \frac{\partial \operatorname{Re}(\alpha(x))}{\partial x} a^{\dagger} a-\hbar \omega_{c} f\left(x_{0}\right) \operatorname{Re}\left(\alpha\left(x_{0}\right)\right) \frac{\partial \omega_{c}}{\partial x} a^{\dagger} a\right] \tag{A4}
\end{array}
$$

## Appendix B

The cavity detuning can be computed using the Bethe-Schwinger cavity perturbation theory as:

$$
\begin{equation*}
\delta \omega=-\frac{\omega_{c}|E(x)|^{2} \operatorname{Re}(\alpha)}{\int_{V c} \epsilon_{c}|E(x)|^{2} d V} \tag{A5}
\end{equation*}
$$

For a particle with electric permittivity $\epsilon_{p}$ in a cavity medium $\epsilon_{m}$, the normalization integral over the cavity volume $V_{c}$ becomes

$$
\begin{equation*}
\int_{V c} \epsilon_{c}|E(x)|^{2} d V=\epsilon_{m} \int_{V c}|E(x)|^{2} d V+\delta \epsilon \int_{V p}|E(x)|^{2} d V \tag{A6}
\end{equation*}
$$

where $\delta \epsilon=\epsilon_{p}-\epsilon_{m}$. Therefore, if we use $\int_{V c}|E(x)|^{2} d V=1$, for a small particle of volume $V_{p}$, we get

$$
\begin{equation*}
\delta \omega=-\frac{\omega_{c}|E(x)|^{2} \operatorname{Re}(\alpha)}{\int_{V c} \epsilon_{c}|E(x)|^{2} d V}=-\frac{\omega_{c}|E(x)|^{2} \operatorname{Re}(\alpha)}{\left[\epsilon_{m}+\delta \epsilon V_{p}|E(x)|^{2}\right]} \tag{A7}
\end{equation*}
$$

and, then

$$
\begin{equation*}
\omega_{c}(x)=\frac{\omega\left[\epsilon_{m}+\delta \epsilon V_{p}|E(x)|^{2}\right]}{|E(x)|^{2} \operatorname{Re}(\alpha)+\left[\epsilon_{m}+\delta \epsilon V_{p}|E(x)|^{2}\right]} \tag{A8}
\end{equation*}
$$

where, in the case of a small spherical dielectric particle

$$
\begin{equation*}
\operatorname{Re}(\alpha)=3 \frac{\epsilon_{m} V_{p}\left(\epsilon_{p}-\epsilon_{m}\right)}{\left(\epsilon_{p}+2 \epsilon_{m}\right)} \tag{A9}
\end{equation*}
$$

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