

Coupled quantum molecular cavity optomechanics with surface plasmon enhancement: comment

SEYED MAHMOUD ASHRAFI,^{1,*}  NARJES TAGHADOMI,¹ ALIREZA BAHRAMPOUR,² AND RASOUL MALEKFAR¹ 

¹Department of Physics, Tarbiat Modares University, Tehran, Iran

²Department of Physics, Sharif University, Tehran, Iran

*Corresponding author: s.ashrafi@modares.ac.ir

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We have found an incorrect formula to estimate the root mean square (rms) amplitude of the molecular vibration in the molecular optomechanics systems reported by J. Liu *et al.* [Photon. Res. 5, 450–456 (2017)]. In contrast with common optomechanical systems, the equilibrium position for molecular optomechanics systems used in the letter cannot be achieved by the equipartition theorem. Here, we achieve the effective temperature of molecules and then provide a corrected formula for the estimation of the rms amplitude of molecular motion. Using defined effective temperature into our new formula, we show that the minimal measurable force is ($F \approx 1.7 \times 10^{-14}$ N), which it is 1 order bigger than the result of the main paper, and it is also in accordance with numerical calculation through the Lindblad master equation. © 2020 Chinese Laser Press

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The model introduced in Ref. [1] by Jian Liu *et al.* consists of two adjacent molecules placed in the hot spot of a plasmonic cavity, which directly interact through dipole–dipole interaction and are also driven by two color laser beams (i.e., a strong pump and a weak probe laser), as shown in Fig. 1. The optomechanical theory has been applied to investigate the coupling behavior of molecules so that the full Hamiltonian in a rotating frame is then given by

$$\begin{aligned} \hat{H} = & \hbar\delta_{\text{pu}}\hat{c}^\dagger\hat{c} + \sum_{i=1,2} \hbar\omega_i\hat{a}_i^\dagger\hat{a}_i - \sum_{i=1,2} \hbar g_i\hat{c}^\dagger\hat{c}(\hat{a}_i^\dagger + \hat{a}_i) \\ & - \hbar\lambda(\hat{a}_1^\dagger\hat{a}_2 + \hat{a}_1\hat{a}_2^\dagger) + i\hbar\Omega_{\text{pu}}(\hat{c}^\dagger - \hat{c}) \\ & + i\hbar\Omega_{\text{pr}}(\hat{c}^\dagger e^{-i\delta t} - \hat{c}e^{i\delta t}), \end{aligned} \quad (1)$$

where \hat{c}, \hat{a}_i are annihilation operators for optical and the vibrational mode of molecule (i), λ is the dipole–dipole coupling constant, $\delta_{\text{pu}} = \omega_c - \omega_{\text{pu}}$ is pump cavity detuning, g_i is optomechanical coupling constant, and $\Omega_{\text{pu}}, \Omega_{\text{pr}}$ are the laser pumping and probing rate, respectively. Using the Lindblad master equation, the dynamical behavior of open whole systems can investigate as follows [2]:

$$\frac{d\hat{\rho}}{dt} = \frac{1}{i\hbar}[\hat{H}, \hat{\rho}] + L_{\hat{c}} + L_{\hat{a}_1} + L_{\hat{a}_2}, \quad (2)$$

where

$$\begin{aligned} L_{\hat{a}_i} = & \frac{\gamma_i}{2}(\bar{n}_i + 1)D_{\hat{a}_i}\{\hat{\rho}\} + \frac{\gamma_i\bar{n}_i}{2}D_{\hat{a}_i^\dagger}\{\hat{\rho}\}, \quad i = 1, 2, \\ L_{\hat{c}} = & \frac{\kappa}{2}D_{\hat{c}}\{\hat{\rho}\}, \end{aligned} \quad (3)$$

where $D_{\hat{A}}\{\hat{\rho}\}$ is defined as the Lindblad super operator,

$$D_{\hat{A}}\{\hat{\rho}\} = 2\hat{A}\hat{\rho}\hat{A}^\dagger - \hat{A}^\dagger\hat{A}\hat{\rho} - \hat{\rho}\hat{A}^\dagger\hat{A}, \quad A = c, a_1, a_2, \quad (4)$$

and $\kappa, \gamma_1, \gamma_2$ are the damping of the plasmon and the first and second molecule, respectively.

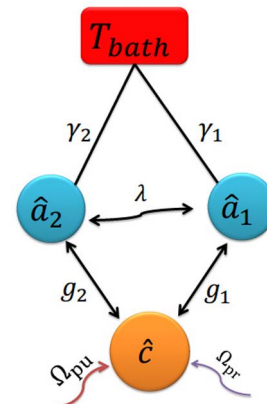


Fig. 1. Schematic of the model and relevant parameters.

Although two molecules are coupled to a common heat bath at temperature T_{bath} (authors had considered room temperature, i.e., $T_{\text{bath}} = 300$ K), optomechanical interaction between molecules and dipole–dipole can modify the temperature of molecules. Hence, we define the effective mean phonon number as

$$n_i^{\text{eff}} = \langle \hat{a}_i^\dagger \hat{a}_i \rangle = \text{tr}(\hat{a}_i^\dagger \hat{a}_i \rho_{\text{ss}}), \quad (5)$$

where ρ_{ss} is the steady-state density operator. As a direct result, the effective temperature of each molecule yields [3]

$$T_i^{\text{eff}} = \frac{\hbar \omega_i}{K_B \log \left(1 + \frac{1}{n_i^{\text{eff}}} \right)}, \quad i = 1, 2. \quad (6)$$

In the symmetric case, when $\omega_1 = \omega_2$ and $g_1 = g_2$, the effective temperature of both molecules is the same, i.e., $T_1^{\text{eff}} = T_2^{\text{eff}}$ (note that two molecules are coupled to a common heat bath). However, for the case $\lambda = 0.2$ THz, we have plotted the effective temperature of molecules as a function of optomechanical coupling constant with two different heat baths (Fig. 2; all parameters are considered similar to the main paper). The

effective temperature of molecule GBT becomes $T_1^{\text{eff}} = T_2^{\text{eff}} \approx 320$ K, when the molecules are coupled to a room temperature heat bath. One can also analytically derive the effective temperature of molecules through adiabatic elimination of plasmon mode, in which an effective Hamiltonian for the two molecules is derived and also two mutually coupled molecules become coupled to an effective common thermal bath [3,4].

Now, we provide a corrected formula for estimation of the root mean square (rms) amplitude of molecular motion and show that the equipartition theorem does not work in molecular optomechanics systems in contrast with the common optomechanical system. The rms amplitude of molecular motion is defined as follows:

$$x_i^{\text{rms}} = \sqrt{\langle \hat{x}_i^2 \rangle} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m} \langle (\hat{a}_i^\dagger + \hat{a}_i)^2 \rangle}, \quad i = 1, 2, \quad (7)$$

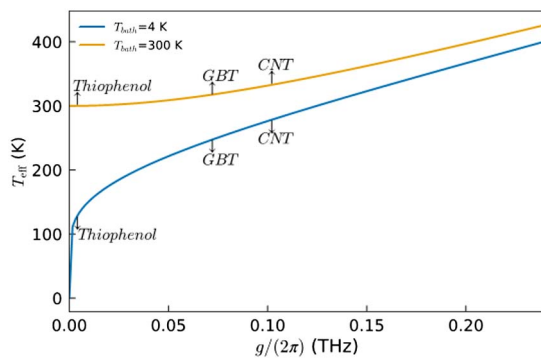


Fig. 2. Effective temperature of molecules as a function of optomechanical coupling constants (notice that all parameters are considered similar to the main paper, i.e., $\frac{\kappa}{2\pi} = 33$ THz, $\frac{\omega_m}{2\pi} = 32.2$ THz, $g/2\pi = 70$ GHz, $\lambda = 0.2$ THz, $\Omega_{\text{pu}}^2 = 0.22$ eV², $\delta = 0$, and we also ignore the probe laser $\Omega_{\text{pr}} = 0$ because it is weak and far detuned from plasmonic cavity center frequency and hence does not play a role in heat transferring).

where m_{eff} is considered as the effective mass of the benzene ring and the atom of sulfur or gold for molecules of thiophenol or GBT ($m_{\text{eff}} \approx 3.6 \times 10^{-26}$ kg) [1]. Using Eq. (5) and the commutation relation, one can easily show that

$$x_i^{\text{rms}} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m} (2n_i^{\text{eff}} + 1)}, \quad i = 1, 2. \quad (8)$$

Then, the rms amplitude can also write as a function of defined effective temperature using Eq. (6),

$$x_i^{\text{rms}} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m} \left[\frac{2}{1 + \exp\left(\frac{\hbar\omega_m}{K_B T_i^{\text{eff}}}\right)} + 1 \right]}, \quad i = 1, 2. \quad (9)$$

After some calculation, the above equation is recast to

$$x_i^{\text{rms}} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m} \coth\left(\frac{\hbar\omega_m}{2K_B T_i^{\text{eff}}}\right)}, \quad i = 1, 2. \quad (10)$$

In the classical limit $K_B T \gg \hbar\omega_m$, the above equation becomes the classical rms amplitude derived by equipartition theorem, in which the average energy is equally shared among all degrees of freedom in the same portion, so we have

$$x_i^{\text{rms}} = \sqrt{\frac{K_B T_i^{\text{eff}}}{m_{\text{eff}}\omega_m^2}}, \quad i = 1, 2 \quad (11)$$

(because of $\coth x \approx \frac{1}{x}$ for $x \ll 1$). But, here system is in the other extreme, i.e.,

$$K_B T \leq \hbar\omega_m. \quad (12)$$

That means the energy of the resonator ($\omega_m/2\pi = 32.2$ THz, or $\hbar\omega_m \approx 133$ meV) is much larger than the room temperature energy ($K_B T_i^{\text{eff}} \approx 28$ meV for GBT), and the high temperature is not valid anymore. Molecular vibrations are indeed “frozen” at room temperature, so that their amplitude of motion is dominated by quantum zero point fluctuations instead of classical thermal fluctuations, which can change the main result of the letter about the minimal measurement force between molecules through a plasmonic cavity. For this point, we plot the minimal measurement force as a function of molecular frequency for three approaches, equipartition theorem (dashed line), numerically by Lindblad master equation in QUTIP package [5] (solid line), and our corrected formula (star dots) in Fig. 3, in which, for the numerical approach, we achieve the rms amplitude as follows:

$$x_i^{\text{rms}} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m} \text{tr}[(\hat{a}_i^\dagger + \hat{a}_i)^2 \rho_{\text{ss}}]}, \quad i = 1, 2. \quad (13)$$

As shown in Fig. 3, the results of our corrected formula are matched with numerical Lindblad approach, which shows that the corrected formula is trustable in quantum regime. In order to clarify the equipartition theorem formula does not work as well, we plot the minimal measurement force for two different effective temperatures (cryogenic and almost room temperature for GBT), which shows a big deviation

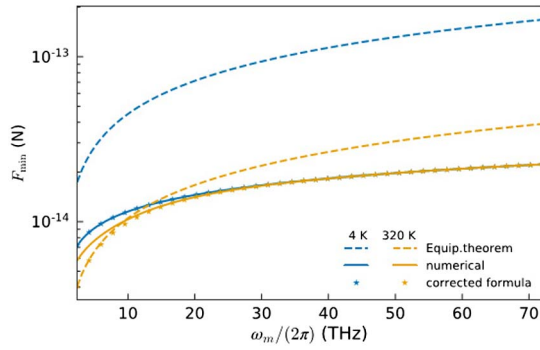


Fig. 3. Comparison between different methods (equipartition theorem, numerical, and corrected formula) for minimal measurable force as a function of molecular frequency, with two different effective temperatures.

especially at high molecular frequency ($\omega_m/2\pi \geq 25$ THz such as GBT) and cryogenic effective temperature. In this situation, $\coth\left(\frac{\hbar\omega_m}{2k_B T_i^{\text{eff}}}\right) \approx 1$, and rms amplitude becomes similar to zero-point motion, i.e.,

$$x_i^{\text{rms}} = \sqrt{\frac{\hbar}{2m_{\text{eff}}\omega_m}}, \tag{14}$$

which shows it is dominated by quantum zero-point fluctuations instead of a thermal environment.

In particular, we achieve the rms of molecules GBT in the main paper at effective temperature ($T_i^{\text{eff}} \approx 320$ K) using our correct formula [Eq. (10)],

$$x_i^{\text{rms}} \approx 0.0027 \text{ nm}. \tag{15}$$

And also the distance between molecules becomes

$$R \approx 0.88 \text{ nm}. \tag{16}$$

Therefore, the minimal measurable force is given as the following relation:

$$F \approx \frac{dV_{\text{int}}}{dR} = \frac{6e^2}{4\pi\epsilon_0 R^4} \langle x_1 \rangle \langle x_2 \rangle \approx 1.7 \times 10^{-14} \text{ N}. \tag{17}$$

One can also investigate the rms amplitude of quantum molecular optomechanical systems through quantum counterparts of the energy partition theorem described by the corresponding frequency probability distributions [6,7].

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