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Laser cooling of an organic quantum well

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Optical cooling of semiconductor devices and microsystems has great potential as a viable solution to thermal management. We theoretically propose a optical scheme to cooling an organic quantum well in the strong coupling regime with quantum master equation. Although there are many modes dwell in the organic quantum well, the single phonon mode that couples to the exciton using a bosonic exciton approximation is only considered. With the exciton-phonon interaction which plays a key role in the system, the ground state cooling can realize. Parameters that dominate the ground state cooling in the system are discussed which may achieve the cooling of the organic film.

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1. Introduction

Over the past few years, the nonlinear optical properties of excitons confined in low dimensional molecular geometries such as crystalline organic superlattices, molecular aggregates, conjugate polymers and molecular monolayers have been focused ones extensive attention [1–3]. These organic systems are characterized by a strong exciton-phonon interaction, which induces large nonlinear optical phenomenon [4,5]. Agarwal has studied the field-induced transparency in such high-density exciton systems neglecting the influence of phonons [6]. Imamoglu et al. analyzed the elementary properties of exciton-polariton lasers which generates coherent optical and matter waves via final state stimulation of excitonphonon scattering [7]. The anomalous optical nonlinearity of polydiacetylene-toluene sulfonate results from phonon-mediated interactions between virtual excitons has also been investigated [8]. Based on the bosonic approximation [2,9], the existence of electromagnetically induced transparency (EIT) due to exciton-phonon coupling in organic quantum wells has demonstrated [4,5], in which the strong exciton-phonon interation plays a key role in the generation of EIT in the organic quantum wells system. EIT [10-11] has been discovered in the atomic vapors, which has leaded to many different applications, particularly in the context of slow light [12,13] and the production of giant nonlinear effects. Therefore optical communications, quantum information processing and quantum nondemolition measurements, as well as for novel acoustical-optical devices might be utilized via EIT. However, many of quantum effects in the system are masked by thermal noise, consequently, the systems should be cooled to their ground states.

Laser cooling of solids and, more specifically, semiconductors quantum wells have attracted the interest of researchers [14-17], and this process carries the promise of developing compact refrigerators for applications in nanoscale electronic and optoelectronic devices. In the past several years, both theoretical and experimental investigations of the cooling of semiconductor have been demonstrated by many groups [18-25], such as Huang and Rupper theoretically investigated the laser cooling of a semiconductor [18-22,], and experimentally reported the laser cooling of a semiconductor [23-25] by several groups. Ground-state cooling of the whole network becomes inefficient as the system size increases, but it is also unnecessary, since usually only a few modes within a small bandwidth are used for transmitting quantum states or quantum information processing. Consequently, laser cooling of a quantum well to its ground state can achieve due to the strong exciton and longitudinal optical phonons (LOPs) coupling in the system.

On the other hand, cooling of rare-earth doped glasses and crystals have also been investigated theoretically and experimentally in the past years [26-30], and this approach to laser cooling of solids permits an increase in the efficiency of the cooling cycle, as well as an acceleration of the cooling process. Moreover, cooling of mesoscopic mechanical resonators represents a primary concern in cavity optomechanics [31,32], and the mechanical resonators can reach to its ground state via radiation pressure in such optomechanics system have also been demonstrated [33,34].

In the present work, we theoretically demonstrate an organic quantum well can be cooled to its ground state with the quantum master equation. Using the bosonic exciton approximation, we only consider a single mode in the organic quantum well system driven by an external field. The ground state cooling is manipulated by the strong exciton-LOP interaction, in which the exciton behaves as an optical cavity and the phonon like a mechanical resonator analogy in cavity optomechanics system. At high phonon frequency and low initial bath temperature T, the phonon can be cooled to its ground state.

2. Theory

We consider an organic quantum well which has an ideal cubic lattice with N lattice sites and contains only one molecular layer where N identical two-level molecules distribute into the lattice sites. The system includes the interaction of excitons and phonons with an external radiation fields. Assuming only existing a one-phonon mode and keeping only the linear exciton-phonon interaction, we obtain the Hamiltonian of the system in the rotation wave approximation [4,5,35]

$$\begin{aligned} H &= -\hbar\Delta_0 S^z + \hbar\omega_{ph} b^\dagger b \\ &+ \hbar g S^z (b + b^\dagger) + \hbar\Omega_0 (S^- + S^\dagger) \end{aligned} \tag{1}$$

The first term on the right of Eq. (1) is the exciton energies, and $\Delta_0 = \omega_c - \omega_{ex}$ is the detuning between the control field and the exciton. $S^z, S^\dagger$ and S^- are the collective operators with the form $S^z = \sum_{j=1}^{N} s_j^z$ and $S^\pm = \sum_{j=1}^{N} s_j^\pm$, where $s_j^z = s_j^+ s_j^- - 1/2$ and $s_j^\pm = s_j^x \pm i s_j^y$ are pesudospin operators with the spin commutation relations $\left[s_j^z, s_j^\pm\right] = \pm s_j^\pm$ and $\left[s_j^+, s_j^-\right] = 2 s_j^z$. The second term is the phonon energies with phonon frequency ω_{ph}. The third term gives the coupling between exciton and phonon with coupling constant g. The last term describes interaction between exciton and external radiation fields. $\Omega_0 = \mu_0 E_c / \hbar$ is the Rabi frequency of the control field, E_c is related to the power of the control field, and μ_0 is the electric dipole moment of the exciton. In the case of low excitation and neglecting the intermolecular interactions, the collective behaviour of many mlecules can be described by a bosonic exciton [2,9]. Introducing the bosonic approximation and defining $a = S^- / \sqrt{N}$ and $a^\dagger = S^+ / \sqrt{N}$ with $\left[a, a^\dagger\right] = 1$, the Hamiltonian of Eq. (1) becomes

$$\begin{aligned} H &= -\hbar\Delta_0 a^\dagger a + \hbar\omega_{ph} b^\dagger b \\ &+ \hbar g a^\dagger a (b + b^\dagger) + \hbar\Omega (a^\dagger + a) \end{aligned} \tag{2}$$

where $\Omega = \sqrt{N}\Omega_0 = \mu E_c / \hbar$ and $\mu = \sqrt{N}\mu_0$.
For strong driving, the Hamiltonian can be linearized with $a \to a + \alpha$ and $b \to b + \beta$, which means a and b are separated into the fluctuations and its mean values $\alpha = \langle a \rangle$ and $\beta = \langle b \rangle$. Neglecting the nonlinear terms and yielding the Hamiltonian

$$\begin{aligned} H_{eff} &= -\hbar\Delta a^\dagger a + \hbar\omega_{ph} b^\dagger b \\ &+ \hbar g (G a^\dagger + G^* a)(b + b^+) \end{aligned} \tag{3}$$

where $\Delta = \Delta_0 + 2|G|^2 / \omega_{ph}$ is the exciton-phonon coupling modified detuning, and $G = \alpha g$ describes the linear coupling strength. Taking the dissipations into consideration, the system is governed by the quantum master equation $\dot{\rho} = i\left[\rho, H_{eff}\right] + \Gamma D[a]\rho + \gamma_{ph}(n_{th}+1) D[b]\rho + \gamma_{ph} n_{th} D\left[b^\dagger\right]\rho$, where $D[\hat{o}]\rho = \hat{o}\rho\hat{o}^\dagger - (\hat{o}^\dagger\hat{o}\rho + \rho\hat{o}^\dagger\hat{o})/2$ denotes the Liouvillian in Lindblad form for operator $\hat{o}$. Γ and γ_{ph} are phenomenological exciton dephasing and phonon-decay rates, respectively, and $n_{th} = \left[\exp(\hbar\omega_{ph}/k_b T) - 1\right]^{-1}$ are occupation number of the phonon related to the bath temperature T.

Starting from the master equation, we obtain a set of differential equations for the mean values of the second-order moments $N_b = \langle b^\dagger b \rangle$, $\langle a^\dagger a \rangle$, $\langle a^\dagger b \rangle$, $\langle ab \rangle$, $\langle a^2 \rangle$, $\langle b^2 \rangle$. In the steady state we obtain the average phonon occupancy [36–38]

$$\bar{N}_{ph} = \frac{\gamma_{ph}(4|G|^2 + \Gamma^2)}{4|G|^2(\Gamma + \gamma_{ph})} n_{th} + \frac{\Gamma^2 + 8|G|^2}{16(\omega_{ph}^2 - 4|G|^2)} \tag{4}$$

where the first term is the classical cooling limit which related to n_{th} and the second term originates from the external radiation fields exciting on the exciton that induces the so called quantum backaction in cavity optomechanics systems. Thus, the final effective temperature T_{eff} of phonon mode can be expressed as $T_{eff} = \omega_{ph} / \left[k_B \ln(1/\bar{N}_{ph} + 1)\right]$.

3. Numberical results and discussions

To illustrate the numerical results, we consider the realistic parameters of the organic quantum well system as follows [5,8]: $\omega_{ph} = 0.2eV$, $\omega_{ex} = 0.2eV$, $\gamma_{ph} = 2meV$, $g = 0.1eV$, $\Gamma = 50meV$. Laser cooling of semiconductor quantum well has been investigated [14,15,39] and the progress of cooling experiences three steps as shown in Fig. 1(a): (a) A laser beam illuminates the material and creates cold electron-hole pairs, (b) the pairs experience energy exchange through inelastic collision and thereby absorb thermal phonon vibrations, and then (c) the pairs convert into blue shifted, incoherent photons through radiative recombination and carry heat away from the material. As a result, the material is cooled down by the laser beam.

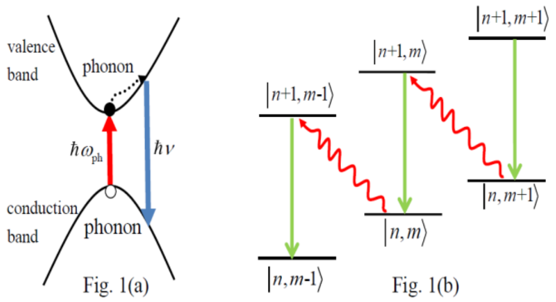

Fig. 1. (a) Schematic of luminescence up-conversion for laser cooling of semiconductors. (b) Level diagram of the cooling process in the organic quamtum well system. $|n,m\rangle$ denotes the state of n photons and m phonons in the displaced frame. The solid curves with arrows correspond to the cooling processes

Here, we adopt an all-quantum scheme to elaborate the cooling progress of the organic quantum well, where the strong exciton-phonon interaction plays a key role in the system. The quantum scheme of cooling is very different from the previous ones [14,15,39]. We diagram the schematic of the cooling process in the strong couple exciton-phonon system (see Fig. 1b). The exciton state $|n\rangle$ and phonon state $|m\rangle$ form the coupled state $|n,m\rangle$. When the system is excited by a red-detuned pump field where $\Delta=\omega_m$, the progress of $|n,m\rangle\to|n+1,m-1\rangle$ is resonantly enhanced. A subsequent decay from the $|n+1\rangle$ state to the $|n\rangle$ state reduces the energy of the phonon by one quanta. This cooling process is described by the state transition $|n,m\rangle\to|n+1,m-1\rangle\to|n,m-1\rangle$ as shown in Fig. 1(b). A series of cycles of this process leads to the cooling of the system. The inverse processes leads to heating which are not shown in Fig. 1(b). In fact, when exerting an external radiation fields in the organic quantum well system, the function of excition in quantum well behave as an optical cavity field in cavity optomechanics system, where the mechanical resonators can be cooling to its ground via radiation pressure [33,34]. Therefore, the organic quantum well can also be reach to its ground with an external light fields.

The organic quantum well is multimode system, ground-state cooling of the whole system is inefficient, and it is also unnecessary, because usually only a few modes or a signal mode are used for transmitting quantum states or quantum information processing. Besides, only the exciton and LOP have the strong coupling, can the ground state cooling be realized, we therefore only consider a single mode with bosonic approximation [2,9] in the organic quantum well system. We first consider the bath temperature is T = 300K, the organic quantum well can be cooled below 25K under a strong external radiation field. When decreasing the bath temperature (such as T = 100K), the final effective temperature T_{eff} will decrease significantly as shown in Fig. 2. If we further decrease the bath temperature and considering the bath temperature approaches 1K, the organic quantum well can reach to its ground state, i.e., the phonon occupancy number is below 1 as shown in the insert of Fig. 2.

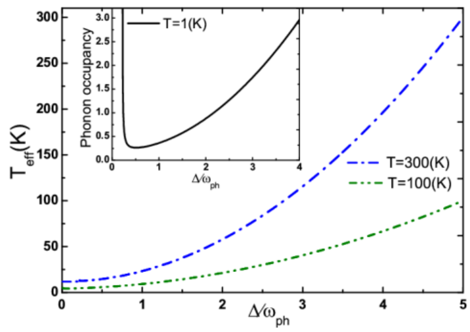

Fig. 2. The final effective temperature T_{eff} as a function of the effective detuning Δ/ω_{ph} with two bath temperature T=300K and T=100K. The insert shows the steady state phonon occupancy number at the bath temperatures T=1K. The other parameters used are $\omega_{ph}=0.2eV$, $\omega_{ex}=0.2eV$, $\gamma_{ph}=2meV$, $g=0.1eV$, and $\Gamma=50meV$

In Fig. 3, we further consider the steady state phonon occupancy number as function of the effective detuning Δ with several different phonon frequencies ω_{ph} under the bath temperature approaches 1K. It is obvious that the phonon occupancy number can below 1, i.e., the ground-state cooling of the organic quantum well can achieve easily at low bath temperature. In addition, compared with the three phonon frequencies $\omega_{ph}=0.20eV$, $\omega_{ph}=0.25eV$, and $\omega_{ph}=0.30eV$, we obtain that high phonon frequency results in better cooling. This behavior of phonon vibration likes phonon cavity and high phonon frequency induces the high phonon cavity quality factor which leads to better cooling. Therefore, there are two key factors that are the bath temperatures and phonon frequency will dominate the cooling of the organic quantum well. The low bath temperatures and high phonon frequency will leads to better cooling.

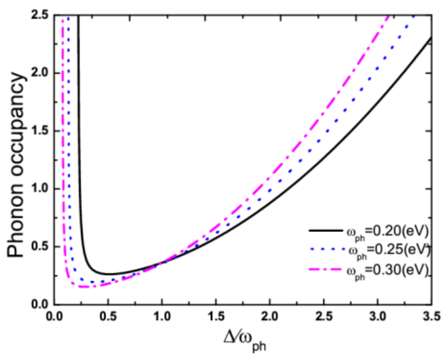

Fig. 3. Curves for the steady state phonon occupancy number versus the effective detuning Δ/ω_{ph} with several different phonon frequency ω_{ph} under the bath temperatures T=1K. The other parameters used as in Fig. 2

4. Summary

We have investigated the cooling of a organic quantum well which is pumped by one control field via the quantum master equation using the bosonic exciton approximation. The exciton-phonon interaction play a key role in the organic quantum well, which induces the ground state cooling. With the strong coupling between exciton and high frequency phonon, the ground state cooling can obtain in the system. The cooling of the organic quantum well provides a new way for exploring the quantum regime of the organic quantum well.

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