Heat pumping with optically driven excitons

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We present a theoretical study showing that an optically driven excitonic two-level system in a solid state environment can act as a heat pump by means of repeated phonon emission or absorption events. We derive a master equation for the combined phonon bath and two-level system dynamics and analyze the direction and rate of energy transfer as a function of the externally accessible driving parameters in the coherent control regime. We discover that if the driving laser is detuned from the exciton transition, cooling the phonon environment becomes possible.

In semiconductor quantum dots (QDs) the ground state and the state containing a trapped electron-hole pair (exciton) form a two level system (2LS) which is a popular implementation of a qubit. Unlike their atomic counterparts, such excitonic qubits are intrinsically coupled to the lattice dynamics of the surrounding material \cite{1, 2, 3}. Longitudinal optical (LO) phonons give rise to excitonic polarons \cite{4} and are known to play an important role for ultrafast excitation \cite{5, 6, 7}. However, in the much slower coherent control regime that is so interesting for quantum information processing, optical phonons only contribute negligibly to the dephasing, and the coupling to longitudinal acoustic (LA) phonons via the deformation potential is predicted to be dominant at low temperatures \cite{8, 9, 10}. In recent experimental studies, the influence of LA phonons on the coherent QD dynamics has been measured by the tunnel charge through the QD (proportional to the excitonic population) following optical excitation \cite{11}, and also through the direct coupling of the QD’s dipole to optical modes in microwaves \cite{12, 13}. These studies confirm that a 2LS model with a perturbative treatment of the LA phonon interaction provides an excellent description in this regime.

Previous theoretical studies of the exciton-phonon interaction in the Rabi regime have fully discarded all information about the state of the phonon bath \cite{5, 6, 11, 14}. Here we develop a technique for tracking the number of excitations in the environment, allowing us to analyze the net rate of absorbed or released bath energy, and show that a continuously driven excitonic qubit constitutes a controllable two-way heat pump. Exploiting this cooling effect would help with gaining further experimental insight into the exciton-phonon coupling, crucial for managing decoherence of semiconductor charge qubits, as well as providing an easy preparatory qubit initialization step for quantum information processing.

\textit{Model} - We consider a self-assembled QD (such as InGaAs encased in GaAs substrate) illuminated by a laser beam with frequency $\omega_l$, which is nearly resonant with the crystal ground state to exciton transition. In a frame rotating with the laser frequency and within the rotating wave approximation (RWA), the system is governed by the Hamiltonian ($\hbar = 1$)

$$ H_S = \Delta/2 \sigma_z + \Omega/2 \sigma_x, \quad (1) $$

where the detuning $\Delta$ describes the energy difference between the basis states in the rotating frame, $\Omega$ is the Rabi frequency.

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\includegraphics[width=\textwidth]{figure.png}
\caption{(color online) The driven exciton acts as a heat pump between the phonon and the electromagnetic environment. If the driving is detuned from the exciton transition, the laser-dressed eigenstates (see Ref. \cite{14}) are composed of different amounts of the ground and excited state. Cooling is possible when spontaneous emission repeatedly takes the system into its lower eigenstate, from where it can only absorb phonons.}
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\caption{Heating with optically driven excitons}
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The exciton-phonon interaction term is generically given by \cite{11}

$$ H_I = \sigma_z \sum_q g_q (\hat{a}_q^\dagger + \hat{a}_q), \quad (3) $$

where the $g_q$ are coupling constants. Moving into the interaction picture with respect to $H_S$ and $H_B$ we obtain for the transformed interaction Hamiltonian

$$ \tilde{H}_I(t) = \tilde{\sigma}_z(t) \sum_q g_q (\hat{a}_q e^{i\omega_q t} + \hat{a}_q^\dagger e^{-i\omega_q t}), \quad (4) $$

where $\tilde{\sigma}_z(t)$ denotes the transformed $\sigma_z$ operator.
For monitoring phonon excitations in the QD’s solid state environment, we adapt a technique from the literature on observing single electron charge transport \([15,17]\) by performing a gauge transformation which adds phase markers to the interaction Hamiltonian. Crucially, these do not alter the system’s dynamics. We define the phonon-number specific density matrix, \(\varrho\), as in Ref. \([16]\). The probability of having emitted a phonon-number operator \(\hat{a}_i\) is given by the density matrix of the combined phonon-qubit system and its expectation value, 

\[
\rho_m = \text{tr}_E(P_m \varrho),
\]

where \(\text{tr}_E\) denotes the trace over the phonon modes, \(\varrho\) is the density matrix of the combined phonon-qubit system and \(P_m\) is a projection operator, 

\[
P_m = \frac{1}{2\pi} \int_0^{2\pi} d\lambda \ e^{-im\lambda} \mathcal{E}_\lambda, \quad \mathcal{E}_\lambda = e^{i\lambda \hat{N}},
\]

with the total phonon number operator \(\hat{N} = \sum_m \hat{a}_m^\dagger \hat{a}_m\), and the gauge transformation operator \(\mathcal{E}_\lambda\). The projection operator \(P_m\) projects out all parts of the wavefunction with a phonon number different from \(m\) (for a detailed discussion see Ref. \([15]\)). The probability of having emitted \(m\) phonons into the environment is then obtained by the matrix trace of the phonon-number specific density matrix 

\[
p_m = \text{tr}(\rho_m).
\]

It is convenient to introduce the Fourier transformed density matrix as 

\[
\rho_\lambda = \sum_m e^{im\lambda} \rho_m = \text{tr}(\varrho_\lambda),
\]

with the definition \(\varrho_\lambda = \mathcal{E}_\lambda/2 \mathcal{E}^\dagger_\lambda/2\). This particular choice corresponds to an initial state with a definite phonon number as in Ref. \([15]\). It can be shown that the full density matrix \(\varrho_\lambda\) in the interaction picture obeys the von Neumann equation, 

\[
\dot{\varrho}_\lambda(t) = -i \left( \hat{H}_\lambda(t) \varrho_\lambda(t) - \varrho_\lambda(t) \hat{H}^\dagger_\lambda(t) \right) = \mathcal{L}_\lambda(t) \varrho_\lambda(t),
\]

defining the super-operator \(\mathcal{L}_\lambda(t)\), where the interaction Hamiltonian \(\hat{H}_\lambda = \mathcal{E}_\lambda/2 \hat{H}^\dagger_\lambda/2\) has acquired phase markers on the phonon creation and annihilation operators: 

\[
\hat{H}_\lambda = \hat{\sigma}_z(t) \sum_q \tilde{g}_q \left( e^{-i\frac{\lambda}{2} \hat{a}_q^\dagger \hat{a}_q} e^{i\omega_q t} + e^{i\frac{\lambda}{2} \hat{a}_q^\dagger \hat{a}_q} e^{-i\omega_q t} \right),
\]

written in a shorter notation as \(\hat{H}_\lambda = \hat{\sigma}_z(t) B_\lambda(t)\). The phase factor \(e^{\pi\lambda/2}\) then keeps track of the creation of phonons, while \(e^{\pi\lambda/2}\) tracks annihilation processes.

We proceed along the standard path of deriving a master equation (ME) \([18]\,\text{[13]}\), and obtain an integro-differential equation for the reduced density matrix of the system \(\rho_\lambda\), 

\[
\dot{\rho}_\lambda(t) = \text{tr}_E \int_0^t ds \mathcal{L}_\lambda(t) \mathcal{L}_\lambda(s) \varrho_\lambda(s).
\]

After the Born-Markov approximation \([18]\) the resulting Markovian ME reads 

\[
\dot{\rho}_\lambda(t) = -\frac{1}{\Gamma} \int_0^\infty ds \sum_{i,j} \mathcal{G}_{ij}^\lambda(s) \mathcal{S}_i(t) \mathcal{S}_j(t - s) \rho_\lambda(t),
\]

where we have defined the super operators \(\mathcal{S}_i(t) = \hat{\sigma}_z(t) \mathcal{L}_\lambda(t) \mathcal{S}_i(t - \lambda)\). The probability of having emitted a gauge transformation operator \(\hat{a}_m\) is given by the density matrix of the combined phonon-qubit system and its expectation value, 

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\rho_m = \text{tr}_E(P_m \varrho),
\]

where \(\text{tr}_E\) denotes the trace over the phonon modes, \(\varrho\) is the density matrix of the combined phonon-qubit system and \(P_m\) is a projection operator, 

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P_m = \frac{1}{2\pi} \int_0^{2\pi} d\lambda \ e^{-im\lambda} \mathcal{E}_\lambda, \quad \mathcal{E}_\lambda = e^{i\lambda \hat{N}},
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with the total phonon number operator \(\hat{N} = \sum_m \hat{a}_m^\dagger \hat{a}_m\), and the gauge transformation operator \(\mathcal{E}_\lambda\). The projection operator \(P_m\) projects out all parts of the wavefunction with a phonon number different from \(m\) (for a detailed discussion see Ref. \([15]\)). The probability of having emitted \(m\) phonons into the environment is then obtained by the matrix trace of the phonon-number specific density matrix 

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\[
\hat{H}_\lambda = \hat{\sigma}_z(t) \sum_q \tilde{g}_q \left( e^{-i\frac{\lambda}{2} \hat{a}_q^\dagger \hat{a}_q} e^{i\omega_q t} + e^{i\frac{\lambda}{2} \hat{a}_q^\dagger \hat{a}_q} e^{-i\omega_q t} \right),
\]

written in a shorter notation as \(\hat{H}_\lambda = \hat{\sigma}_z(t) B_\lambda(t)\). The phase factor \(e^{\pi\lambda/2}\) then keeps track of the creation of phonons, while \(e^{\pi\lambda/2}\) tracks annihilation processes.

We proceed along the standard path of deriving a master equation (ME) \([18]\), and obtain an integro-differential equation for the reduced density matrix of the system \(\rho_\lambda\), 

\[
\dot{\rho}_\lambda(t) = \text{tr}_E \int_0^t ds \mathcal{L}_\lambda(t) \mathcal{L}_\lambda(s) \varrho_\lambda(s).
\]
This equation describes the joint phonon-qubit dynamics in the case where the phonon environment is only weakly disturbed from thermal equilibrium by the excitonic qubit.

**Phonon-assisted transitions** - We now apply Eq. (10) to an excitonic qubit that is optically driven by a pulse of constant intensity $\Omega$. The initial state at $t = 0$ is the system ground state with zero excitations in the environment: $\rho_n(0) = \delta_{n,0} \ket{g}\bra{g}$. For simplicity, we define the spectral density phenomenologically: $J(\omega) = \alpha \omega^3 \exp \left(-\omega^2/\omega_0^2\right)$, where $\alpha$ describes the effective electron-phonon coupling strength and $\omega_0$ is the high frequency phonon cut-off. For relatively weak driving with a peak Rabi frequency $\Lambda$ well below both the electron and the hole cut-off, we can neglect the exponential cut-off term altogether. Setting $\alpha = 1/4 \text{ ps}^2$ yields a coupling strength that is consistent with the magnitude of the GaAs deformation potential reported in the literature [6, 20].

The structure of Eq. (10) permits the emission or absorption of no more than a single phonon: The system is initialised in the superposition of system eigenstates. The Lindblad operator $P_A$ induces a transition from $\ket{+}$ to $\ket{-}$ while $P_A^\dagger$ raises population from $\ket{-}$ to $\ket{+}$. Once a decay process has happened, we find the system in the $\ket{-}$ state, meaning it cannot decay again.

Provided the excitation is sufficiently long, the population ratios thus tend to a Boltzmann distribution, as is obvious from the phonon emission rate proportional to $n(\Lambda) + 1$ and the absorption rate proportional to $n(\Lambda)$,

$$\lim_{t \to \infty} \frac{\text{tr}(\rho_0(t)\ket{+}\bra{+})}{\text{tr}(\rho_1(t))} = \lim_{t \to \infty} \frac{\text{tr}(\rho_{-1}(t))}{\text{tr}(\rho_0(t)\ket{-}\bra{-})} = e^{-\beta \Lambda}.$$ 

So far, the only perturbation to the system has been caused by the coupling to the phonon bath, resulting in single phonon emission and absorption. Realistically, other dissipative processes will be present in any physical systems. We shall include these using additional phenomenological noise operators, such as pure dephasing and radiative decay of the exciton. We model these processes with an additional Lindblad dissipator on the right-hand side of Eq. (10),

$$\mathcal{D} \rho = \Gamma \left( L \rho L^\dagger - \frac{1}{2} (L^\dagger L \rho + \rho L^\dagger L) \right),$$

with respective Lindblad operators $L = \sigma_z$ and $L = \sigma_x$, and where $\Gamma$ is the dephasing or decay rate. These operators do not preserve the system’s eigenstates; consequently, under their action, phonon-assisted transitions become possible in any of the $\rho_n$ subspaces. This leads to a dynamic equilibrium, where phonon-assisted transitions keep occurring after the transient (coherent) evolution of $\rho = \sum_n \rho_n$ has subsided. Our theoretical model is well-suited for illustrating this behaviour: Fig. 2 presents the $p_n$ distribution at different points of time for the case of optical decay.

The pure dephasing $\sigma_z$ operator randomises the phase between $\ket{g}$ and $\ket{e}$, thus balancing the population of $\ket{-}$ and $\ket{+}$. Consequently, once the steady state has been reached, phonon emission always occurs with a faster rate then absorption, and the distribution is therefore shifted in the direction of increasing $n$, meaning the average number of emitted phonons increases. On the other hand, the distribution can move in either direction for optical decay from $\ket{e}$ to $\ket{g}$. For $\Delta = 0$, $\ket{g}$ consists of an equal superposition of $\ket{-}$ and $\ket{+}$, while it contains a larger $\ket{-}$ component for $|\Delta| > 0$. Under these latter circumstances, it is possible for phonon absorption to permanently dominate over emission, shifting the distribution in the direction of decreasing $n$, as shown in the right panel of Fig. 3. In this case, thermal energy is removed from the QD’s bulk surroundings and released into the wider environment by spontaneous photon emission (depicted in Fig. 1).

**Heat transfer rate** - To quantify this observation, we consider only the radiative decay operator of Eq. (11) in the following. We proceed by calculating the rate of phonon emission or absorption, which is given by [17]:

$$\langle \dot{n}(t) \rangle = \int \frac{d\lambda}{\Omega^2} \sum_m m p_m \left| \frac{d}{dt} \text{tr}(\rho_\lambda) \right|_{\lambda=0} = 2 \text{ tr} \left[ \Gamma \mathcal{Q} \rho(t) \mathcal{Q}^\dagger - \Gamma \mathcal{Q}^\dagger \rho(t) \mathcal{Q} \right],$$

where $\rho(t)$ is obtained by integrating Eq. (10) inclusive of the Lindblad dissipator (11) with $L = \sigma_-$ and disregarding the indices $m$ of $\rho_m$.

Fig. 2 presents the steady-state value $\langle \dot{n} \rangle_{ss}$ of Eq. (12) as a function of $\Delta$. As expected, net phonon absorption is only possible at finite temperature for off-resonant excitation. It is a natural question to ask at which rate energy is transferred to or from the surroundings of the system. Considering the quantity $\dot{E} = \sqrt{\Omega^2 + \Delta^2} \langle \dot{n} \rangle_{ss}$ shows that the number of absorbed phonons increases for a larger detuning, yet their energy is greater, shifting the $\Delta$ that achieves optimal heat transfer. The inset of Fig. 3 shows that the process is limited by the radiative decay time $\Gamma$ of the system. A saturation only occurs when the spontaneous emission rate becomes faster than that of phonon-mediated transitions, but this regime would require an unrealistic optical lifetime of the order of a picosecond or less.
FIG. 3: (color online) The rate of phonon-induced transitions \( \langle n \rangle \)ss (ps-1) (red) and the rate of energy transfer \( E \) (ps-2) for fixed \( \Delta = 1 \text{ ps}^{-1} \) as a function of \( \Gamma \) at \( T = 10 \) K.

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[21] A small renormalization due to principal value terms in the ME can be absorbed in the free evolution of the qubit.
[22] Using a mass density of \( \mu = 5.3 \times 10^3 \) kg/m\(^3\) and a specific heat of 350 J/(kg K) gives a heat capacity of 1.85 \times 10^{-12} \) J/K for a micrometer cube of GaAs.
[23] This may also require a straightforward substitution of \( J(\omega) \) to account for a modified phonon spectrum.