RESONANT AMPLIFYING OF RABI OSCILLATIONS AND EXCITED STATE DECAY

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Introduction

The non-radiative excited state decay (or transfer) in quantum dots (QD) or molecular systems is one of the most important processes that controls the efficiency of LED (light emitting diodes). It turned out that the interaction of electronic states with local vibrations may in special instances to show similar properties in both semiconductors or chemical physics. Indeed, it was calculated that the rate of excited state decay in GaAs QD is resonantly amplified for the integer ratio of the electronic excitation energy and energy of longitudinal optical phonons [1]. On the other hand, very similar resonant properties have been found for the excited state decay dissipated on a single vibrational model embedded in a heat bath of passive modes [2]. The latter model is a standard model of the energy and charge transfer in chemical physics. In our work we utilize the same model and we show that simultaneously with resonantly amplified excited decay there is also a significant increase of Rabi oscillations of the excited state population. The latter effect corresponds to the so called adiabatic limit, where a strong coherent coupling between the excited and ground electronic states is found.

Theoretical

The excited state population probablity $p_1(t)$ is defined as

$$p_1(t) \equiv \mathrm{Tr}_{\mathrm{vib}}\{\langle 1|\rho(t)|1\rangle\}, \qquad (1)$$

where $\mathrm{Tr}_{\mathrm{vib}}$ means the trace operation over vibrational mode and state $|1\rangle$ denotes the excited electronic state. $\rho(t)$ is the density matrix of two electronic states and a single vibrational mode, and its time dependence is given by the solution.

$$\frac{\partial}{\partial t}\rho(t) = -\frac{\mathrm{i}}{\hbar}[H, \rho(t)] - \mathrm{i}L^{\mathrm{dis}}\rho(t). \qquad (2)$$

In Eq. 2 H denotes the Hamiltonian of the two electronic levels coupled to a single vibrational mode. It is parameterized by the electronic excitation energy ε, Huang-Rhys factor S of the mode and the inter-state coupling $W(Q)$ that is either constant or linear in the vibrational displacement Q. Values of parameters will correspond to the case of inverted regime. The last term in Eq. 2 describes the process of vibrational dissipation influenced by the bilinear single mode-passive modes coupling. We have used the diabatic damping approximation (DDA) [3], which can be utilized in the inverted regime. For simplicity we will parameterize DDA by the rate of vibrational dephasing k, which will be the same for both potential energy surfaces.

Results and discussion

Typical case of the time evolution of the excited state population probability $p_1(t)$, initially excited to the upper PES from the ground one, is shown in Fig. 1 up to times $t = 250/\omega \sim 4.5$ ps. Generally we observe a superposition of the Rabi oscillations with a population decay. The numerically calculated dependences of the population probability $p_1(t)$ are then also fitted by the superposition of a central excited state decay superimposed with two gradually decaying Rabi oscillations. We then arrive at the following approximate relation

$$p_1(t) \approx A_0 + A_1 \exp(-\Gamma_1 t) +$$

$$+ A_2 \exp(-\Gamma_2 t)\cos(\omega_2 t) + A_3 \exp(-\Gamma_3 t)\cos(\omega_3 t) \quad (3)$$

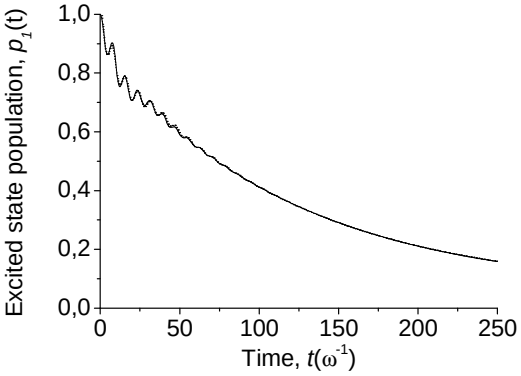


Fig.1 Time dependences of the excited state population decay. The numerically found dependences according Eqs. 1-2 are not distinguished from the fit (3).

Next, we then studied the dependence of the rate of the excited state decay Γ_1 on the ratio $r = \frac{\varepsilon}{\hbar\omega}$ of the electronic excitation energy ε and vibrational quantum $\hbar\omega$. For the case of linear inter-state coupling and zero value of the Huang-Rhys factor we have found dependences as shown in Fig.2

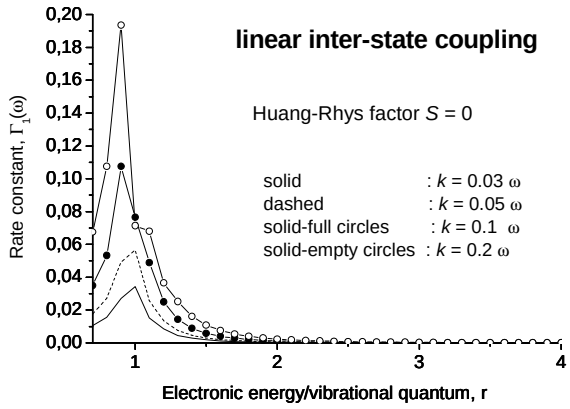
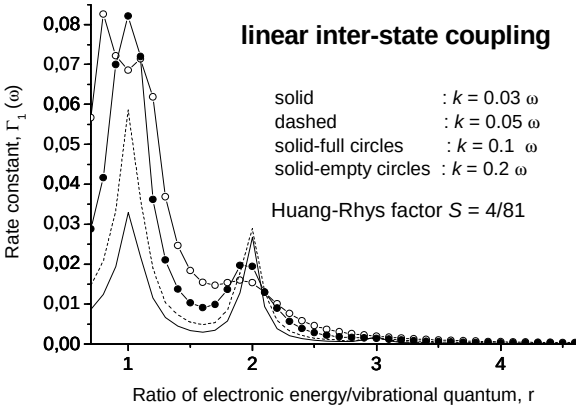


Fig. 10 Dependence of the rate of electronic excited state decay Γ_1 on the ratio $r = \varepsilon/(\hbar\omega)$

In this case we observe a significant resonant amplifying when the excitation energy ε equals to that of vibrational quantum $\hbar\omega$. Moreover, for the resonant condition the decay rate Γ_1 equals to the rate of vibrational dephasing *k*. For non-zero value of the Huang-Rhys factor *S* we find dependences as in Fig. 3. We observe a significant increase of the second resonant peak even for small value of the Huang-Rhys factor *S*. For the case of first resonance we find $\Gamma_1 \approx k$.



Conclusion

Above mentioned resonant peaks were strongly correlated with the competition of effective resonant inter-state coupling and rate of vibrational dephasing. Simultaneously with the excited state decay we have found a pronounced increase of Rabi oscillations. We could explain the effects beyond a finite order perturbation theory.

References

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