

Interplay of excitonic and phonon-mediated Stark effects in quantum wells

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We calculate the pump-probe spectrum of a quasi-2D semiconductor, when the detuning between the pump and probe matches the optical phonon frequency of the material and the probe is tuned across the resonance. The interplay of phonon-mediated and phase space filling nonlinearities is discussed. We show that the relative contribution of the former mechanism should increase at lower temperatures.

1. Introduction

The nonlinear optical properties of Wannier excitons in semiconductors have been extensively studied in the past year [1,2]. Numerous optoelectronic effects were discussed; among them an important one is phase-space filling [1,2], which attributes the nonlinear optical properties of excitons to the Fermi-particle statistics of electrons and holes. Another type of nonlinearity is related to phonon-mediated interaction between excitons. This type of interaction was found to be particularly important in PTS polydiacetylene [3,4], which is a 1D semiconductor-type optically active material. A good agreement between theoretical calculations and experiments was reached on the basis of a model which neglects the phase space filling contribution to excitonic nonlinearity [3]. The interplay between phase-space filling and phonon-mediated nonlinearity in other materials such as GaAs–AlGaAs still remains an open question [1].

In this paper we study a microscopic model of a two-band semiconductor coupled to acoustic and optical phonons. We derive equations of motion for two-point equal-time electronic variables and for the displacement of optical phonons,

based on a coherent state approximation for phonons, and a generalized coherent-state ansatz described in ref. [5]. Temperature is incorporated phenomenologically using a phonon self-scattering rate. We conclude that at high temperatures, the phonon-mediated nonlinearity should be negligible, and we recover the picture of optical nonlinearity of GaAs described in refs. [1,2]. However, at very low temperatures when phonon self-scattering can be neglected the phonon-mediated nonlinearity may become dominant.

2. Two-band semiconductor coupled to phonons

The two-band model Hamiltonian of a semiconductor interacting with an external electromagnetic field and with phonons, has the following form (the electronic part of the Hamiltonian was discussed in ref. [5]);

$$\begin{aligned} H = & \int d\mathbf{r} \hat{\Psi}^\dagger(\mathbf{r}) \\ & \times \left(\frac{\hat{\mathbf{p}}^2}{2m} + V_B(\mathbf{r}) + e\mathbf{r} \cdot E_T(\mathbf{r}, t) \right) \hat{\Psi}(\mathbf{r}) \\ & + \int d\mathbf{r}_1 d\mathbf{r}_2 \hat{\Psi}^\dagger(\mathbf{r}_1) \hat{\Psi}^\dagger(\mathbf{r}_2) \hat{\Psi}(\mathbf{r}_2) \hat{\Psi}(\mathbf{r}_1) \\ & \times V_c(\mathbf{r}_1 - \mathbf{r}_2) \end{aligned}$$

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$$\begin{aligned}
& + \int d\mathbf{r} \sum_{\mathbf{q}} \hat{\Psi}^\dagger(\mathbf{r}) \hat{\Psi}(\mathbf{r}) \left\{ (\hat{b}_{\mathbf{q}} + \hat{b}_{-\mathbf{q}}^\dagger) \chi_{\mathbf{q}}^{(1)} \right. \\
& + (\hat{B}_{\mathbf{q}} + \hat{B}_{-\mathbf{q}}^\dagger) \chi_{\mathbf{q}}^{(2)} \left. \right\} e^{i\mathbf{q}\mathbf{r}} \\
& + \sum_{\mathbf{q}} \left(\Omega_{\mathbf{q}}^{(1)} \hat{b}_{\mathbf{q}}^\dagger \hat{b}_{\mathbf{q}} + \Omega_{\mathbf{q}}^{(2)} \hat{B}_{\mathbf{q}}^\dagger \hat{B}_{\mathbf{q}} \right). \quad (1)
\end{aligned}$$

Here $\hat{b}_{\mathbf{q}}^\dagger$ ($\hat{B}_{\mathbf{q}}^\dagger$) denotes the creation operator of an acoustic (optical) phonon with momentum \mathbf{q} . They satisfy the Bose commutation relations $[\hat{b}_{\mathbf{q}}, \hat{b}_{\mathbf{q}'}^\dagger] = \delta(\mathbf{q} - \mathbf{q}')$, $[\hat{B}_{\mathbf{q}}, \hat{B}_{\mathbf{q}'}^\dagger] = \delta(\mathbf{q} - \mathbf{q}')$. $\chi_{\mathbf{q}}^{(1)}$ and $\chi_{\mathbf{q}}^{(2)}$ denote the (momentum dependent) coupling between acoustic and optical phonons and electrons. We adopt the same notation of ref. [5] i.e. $V_{\text{c}}(\mathbf{r})$ is the instantaneous Coulomb potential, $V_{\text{B}}(\mathbf{r})$ is the periodic lattice potential, $E_{\text{T}}(\mathbf{r})$ is the transverse electric field and $\hat{\Psi}(\mathbf{r})$ is the electronic field operator, which using the Bloch basis set has the form;

$$\hat{\psi}(\mathbf{r}) = \sum_{\mathbf{k}} (\hat{c}_{\mathbf{k}} \phi_{\text{c}\mathbf{k}}(\mathbf{r}) + \hat{d}_{\mathbf{k}}^\dagger \phi_{\text{v}\mathbf{k}}(\mathbf{r})), \quad (2a)$$

$$\phi_{\alpha\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}} u_{\alpha}(\mathbf{r}). \quad \alpha = \text{c, v} \quad (2b)$$

Here $\phi_{\text{c}\mathbf{k}}(\mathbf{r})$ ($\phi_{\text{v}\mathbf{k}}(\mathbf{r})$) are the Bloch functions, $U_{\alpha\mathbf{k}}(\mathbf{r})$ denotes the periodic part of the Bloch function and $\hat{c}_{\mathbf{k}}$ and $\hat{d}_{\mathbf{k}}^\dagger$ denotes annihilation operators for electrons and holes with momentum \mathbf{k} ($-\mathbf{k}$). These operators satisfy $\{\hat{c}_{\mathbf{k}}, \hat{c}_{\mathbf{k}'}\} = 2\delta(\mathbf{k} - \mathbf{k}')$ and $\{\hat{d}_{\mathbf{k}}^\dagger, \hat{d}_{\mathbf{k}'}\} = 2\delta(\mathbf{k} - \mathbf{k}')$.

Using the definitions we can rewrite the Hamiltonian (eq. (1)) in the form:

$$\begin{aligned}
H = & \sum_{\mathbf{k}} (\epsilon_{\text{c}}(\mathbf{k}) \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} + \epsilon_{\text{v}}(\mathbf{k}) \hat{d}_{\mathbf{k}}^\dagger \hat{d}_{\mathbf{k}}) \\
& + \sum_{\mathbf{k}\mathbf{q}} \tilde{d}_{\text{cv}} (E_{\text{T}}(\mathbf{q}, t) \hat{c}_{\mathbf{k}+\mathbf{q}} \hat{d}_{-\mathbf{k}} + \text{h.c.}) \\
& + \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}} \tilde{V}_{\text{c}} \left(\hat{c}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2}^\dagger \hat{c}_{\mathbf{k}_2+\mathbf{q}} \hat{c}_{\mathbf{k}_1-\mathbf{q}} \right. \\
& + \hat{d}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2}^\dagger \hat{d}_{\mathbf{k}_2+\mathbf{q}} \hat{d}_{\mathbf{k}_1-\mathbf{q}} \\
& \left. - 2 \hat{c}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2}^\dagger \hat{d}_{\mathbf{k}_2+\mathbf{q}} \hat{c}_{\mathbf{k}_1-\mathbf{q}} \right) \\
& + \sum_{\mathbf{k}\mathbf{q}} \left(\hat{c}_{\mathbf{k}_2+\mathbf{q}} \hat{c}_{\mathbf{k}} - \hat{d}_{\mathbf{k}_2+\mathbf{q}} \hat{d}_{\mathbf{k}} \right) \left[(\hat{b}_{\mathbf{q}} + \hat{b}_{-\mathbf{q}}^\dagger) \tilde{\chi}_{\mathbf{q}}^{(1)} \right.
\end{aligned}$$

$$\begin{aligned}
& \left. + (\hat{B}_{\mathbf{q}} + \hat{B}_{-\mathbf{q}}^\dagger) \tilde{\chi}_{\mathbf{q}}^{(2)} \right] \\
& + \sum_{\mathbf{q}} \left(\Omega_{\mathbf{q}}^{(1)} \hat{b}_{\mathbf{q}}^\dagger \hat{b}_{\mathbf{q}} + \Omega_{\mathbf{q}}^{(2)} \hat{B}_{\mathbf{q}}^\dagger \hat{B}_{\mathbf{q}} \right), \quad (3)
\end{aligned}$$

where the notation $\tilde{f}(\mathbf{q}) \equiv \int e^{i\mathbf{q}\mathbf{r}} f(\mathbf{r}) d\mathbf{r}$ was used. (\tilde{f} stands e.g. for $\tilde{V}_{\mathbf{q}} \equiv \tilde{V}_{\text{c}}$, $\tilde{\chi}_{\mathbf{q}}^{(1)}$ and $\tilde{\chi}_{\mathbf{q}}^{(2)}$).

In the following we will consider a near-resonant excitation of 1s Wannier excitons. We will assume that the electronic correlation function

$$G(\mathbf{r}_1, \mathbf{r}_2, t) \equiv \langle \hat{\Psi}(\mathbf{r}_1, t) \hat{\Psi}^\dagger(\mathbf{r}_2, t) \rangle \quad (4a)$$

does not depend on $\mathbf{R} \equiv (\mathbf{r}_1 + \mathbf{r}_2)/2$ but only on $\mathbf{r} \equiv \mathbf{r}_1 - \mathbf{r}_2$ (homogeneous excitation). We will further ignore the degeneracy of the hole-band and of the intraband dipole matrix element. The optical polarization is then determined by the excitonic polarization [2]:

$$P_{1s}(t) = \sum_{\mathbf{k}} \tilde{\phi}_{1s}(\mathbf{k}) \langle \hat{c}_{\mathbf{k}} \hat{d}_{\mathbf{k}} \rangle, \quad (4b)$$

where $\tilde{\phi}_{1s}(\mathbf{k})$ is the hydrogenic wave function in the \mathbf{k} -space. We shall explore the dynamics of the system by using the Heisenberg equations of motion originating from the Hamiltonian (eq. (1)), for the following operators

$$\hat{P}_{1s}(t) \equiv \sum_{\mathbf{k}} \tilde{\phi}_{2s}(\mathbf{k}) \langle \hat{c}_{\mathbf{k}} \hat{d}_{-\mathbf{k}} \rangle, \quad (5a)$$

$$\hat{n}_{1s}(t) \equiv \frac{1}{2} \sum_{\mathbf{k}} \tilde{\phi}_{2s}(\mathbf{k}) (\hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} + \hat{d}_{\mathbf{k}}^\dagger \hat{d}_{\mathbf{k}}), \quad (5b)$$

$$\hat{Q}_{\mathbf{q}}^{(1)}(t) \equiv \hat{B}_{\mathbf{q}} + \hat{B}_{-\mathbf{q}}^\dagger \quad (5c)$$

and

$$\hat{Q}_{\mathbf{q}}^{(2)}(t) \equiv \hat{b}_{\mathbf{q}} + \hat{b}_{-\mathbf{q}}^\dagger. \quad (5d)$$

We will further make the following ansatz for the wave function of the system [5]

$$\begin{aligned}
|\Psi_{\Lambda}(t)\rangle = & \prod_{\mathbf{k}\mathbf{q}\tilde{\mathbf{q}}} N_{\mathbf{k}} \exp \left(\eta_1(\mathbf{k}, t) \hat{c}_{\mathbf{k}}^\dagger \hat{d}_{\mathbf{k}}^\dagger + \eta_2(\mathbf{q}, t) \hat{b}_{\mathbf{q}}^\dagger \right. \\
& \left. + \eta_3(\tilde{\mathbf{q}}, t) \hat{B}_{\tilde{\mathbf{q}}}^\dagger \right) |\Omega\rangle, \quad (6)
\end{aligned}$$

where $|\Omega\rangle$ is the ground (vacuum) state and

$$N_{\mathbf{k}}^2(t) = \sqrt{1 - 4\eta_1^2(\mathbf{k}, t)} + 1. \quad (7)$$

The ansatz (6) allows us to factorize the Heisenberg equations derived from eq. (1) and obtain a closed set of equations for the expectation values of the operators defined in eqs. (5a)–(5d). Hereafter we consider only a single optical-phonon mode with coordinate Q , momentum q_0 and damping rate (γ_{ss}). The resulting equations of motion are;

$$\begin{aligned} & \left(i \frac{\partial}{\partial t} - \epsilon_{1s} + i \frac{1}{T_2} \right) P_{1s}(t) \\ &= \tilde{d}_{cv} \frac{E_T(t)}{2} (2n_{1s}(t) - 1) + \lambda Q(t) P_{1s}(t), \end{aligned} \quad (8a)$$

$$\begin{aligned} & \left(i \frac{\partial}{\partial t} + i \frac{1}{T_1} \right) n_{1s}(t) = E_T(t) (\tilde{d}_{cv} P_{1s}(t) \\ & \quad - \tilde{d}_{cv}^* P_{1s}^*(t)), \end{aligned} \quad (8b)$$

$$\frac{\partial^2}{\partial t^2} Q(t) + \gamma_{ss} \frac{\partial}{\partial t} Q(t) + \Omega_0^2 Q(t) = 2\Omega_0 \lambda n_{1s}(t), \quad (8c)$$

where $\lambda \equiv \chi_{q_0}^{(1)}$, $\Omega_0 \equiv \Omega_{q_0}^{(1)}$ and [5]

$$\tilde{d}_{cv} = \int_{-R_{cv}/2}^{R_{cv}/2} d\rho U_c(\rho) U_v(\rho) \rho. \quad (9)$$

It should be noted that the weak-field nonlinear response of this model system can be described using the third order nonlinear susceptibility $\chi^{(3)}$ which can be calculated exactly for the present Brownian-oscillator model [6].

3. The impact of coupling to optical phonons on pump-probe spectroscopy

We have solved eqs. (8a)–(8c) numerically using the Bulirsch–Stoër integration technique. We model the electric field as a sum of two pulses;

$$E(t) = E_1(t) + E_2(t), \quad (10)$$

where $E_1(t)$ denotes the pump pulse and $E_2(t)$ denotes the probe pulse. Similar to ref. [8], we find the differential spectrum $D(\omega)$ by first solving eqs. (8) for $E_1(t) = 0$ and then for $E_1(t) \neq 0$.

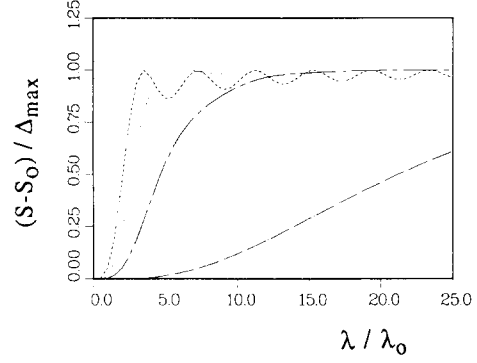


Fig. 1. The first moment of the absorption profile is plotted as a function of the electron phonon coupling constant λ for various values of the optical phonon self-scattering rate. $T_2 = 1$ ns, $T_1 = 3$ ns, $\sigma_1 = 200$ fs, $\sigma_2 = 50$ fs, $\Omega_0 = 5 \times 10^{13}$ Hz, $\Delta_1 = 0$, $\Delta_2 = 5 \times 10^{13}$ Hz, $E_1^0 \tilde{d}_{cv} \sigma_1 = 1$. The various plots correspond to: dash: $\gamma_{ss} = 0$; dot: $\gamma_{ss} T_2 = 10^{-2}$; long and short dash: $\gamma_{ss} T_2 = 1$; dash-dot: $\gamma_{ss} T_2 = 10$, and the coupling constant $\lambda_0 T_2 = 7$.

We then calculated the first moment of the differential absorption spectrum;

$$S = \int d\omega (\omega - \epsilon_{1s}) D(\omega) / \int d\omega D(\omega), \quad (11a)$$

where

$$D(\omega) \equiv \int dt e^{-i\omega t} P_{1s}(t). \quad (11b)$$

We have calculated S for various values of the electron–phonon coupling and the electron self-scattering rate. We used the electronic parameters ϵ_{1s} and \tilde{d}_{cv} and the relaxation rates T_1 and T_2 used in ref. [7]. The pump and probe pulses were assumed to have Gaussian shapes;

$$E_j(t) = E_j^0 \exp(-i\Delta_j t - t^2/\sigma_j^2) \quad j = 1, 2, \quad (12)$$

where σ_j denote the durations and Δ_j denote the detunings of the pump and the probe pulses from the excitonic resonance. The differential absorption shift is plotted for various values of the parameters λ and γ_{ss} in fig. 1. S_0 denotes the shift for $\lambda = 0$. For small values of the phonon self-scattering rate, the shift of the absorption spectrum is very sensitive to the electron–phonon coupling constant. For a large self-scattering rate ($\gamma_{ss} T_2 = 10$) the absorption spectrum shift is much less sensitive to the coupling with optical phonons.

We therefore expect the contribution of electron–phonon coupling to the pump-probe signal to decrease with temperature.

4. Conclusions

We have derived equations of motion describing optically driven Wannier excitons interacting with optical and acoustic phonons in quantum wells. Our results generalize previously discussed theories of phonon-assisted excitonic nonlinearity [6] and optical nonlinearity due to interaction between excitons and optical phonons in polydiacetylene [3,4]. Employing these equations for the two-point electronic correlation function describing the optical response of Wannier excitons, we have studied the impact of interaction of excitons with optical phonons. We find that at high temperatures, the phonon-self-scattering should suppress the excitation of the optical phonon modes, and the approach discussed in ref. [7] where optical phonons are neglected, is justified. We expect, however, that for sufficiently low temperatures, the nonlinearity mediated by optical phonons could become significant.

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