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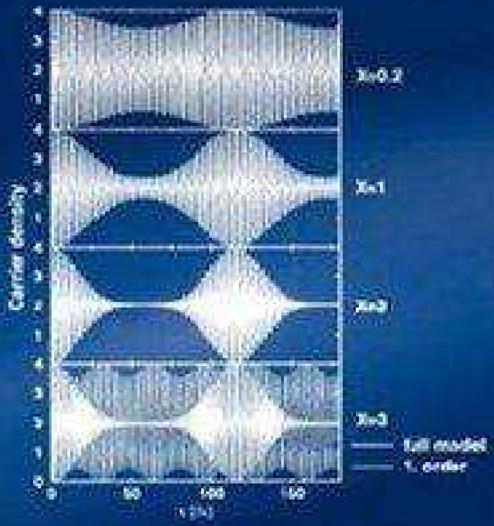

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COHERENT CONTROL OF HEAVY-LIGHT HOLE AND PHONON QUANTUM BEATS

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Abstract. The control of quantum beats by two phase-locked optical pulses is analyzed theoretically. We compare the case of heavy-light hole quantum beats as an example of a purely electronic coherence with the case of phonon quantum beats which are a characteristic feature of electron-phonon quantum kinetics. The maxima and minima of the beat amplitudes can be understood by the same arguments. For the case of phonon quantum beats an exacly solvable model is presented which allows us to analyze the role of the electron-phonon coupling constant.

1. Introduction

Coherent dynamics in semiconductors relies on the fact that a quantum mechanical system is not completely specified by the amplitude of the wave function related to an occupation probability, but also by its phase. In atomic physics such coherent experiments have a long tradition, in semiconductors, due to the short dephasing times, they have been possible only when sufficiently short laser pulses became available. Among the many types of coherent experiments one of the most direct ways to show the wave-like behavior are coherent control experiments which are based on the phenomenon of constructive or destructive interference. Essentially two different classes of such experiments have been performed: The excitation by two simultaneous pulses with different frequencies allows one to control the final state of the system which is used, e.g., for the coherent control of photocurrent [1] or chemical reactions [2]. The excitation by two temporally non-overlapping pulses, on the other hand, gives a direct information on the phase memory introduced by the first pulse. Examples of this latter case are coherent control of exciton density, spin, and heavy-light hole (HL) beats [3, 4, 5], of THz emission due to quantum beats in asymmetric double quantum wells [6, 7], and of phonon quantum beats [8, 9]. In this contribution we will concentrate on the coherent control of quantum beats by comparing the case of HL beats which can be understood on the basis of the semiconductor Bloch equations (SBE) with the case of phonon quantum beats, which require a quantum kinetic treatment of carrier-phonon interaction. Besides a full quantum kinetic semiconductor model we investigate an analytically solvable model which allows us to study the effects of a stronger electron-phonon coupling.

2. Heavy-light hole quantum beats

The theoretical description of coherent dynamics in a semiconductor including heavy holes (HH) and light holes (LH) is based on the multiband SBE. The basic variables are the elements of the single particle density matrix $f^e_{\mathbf{k}} = \langle c^\dagger_{\mathbf{k}} c_{\mathbf{k}} \rangle$, $f^h_{ij,\mathbf{k}} = \langle d^\dagger_{i,\mathbf{k}} d_{j,\mathbf{k}} \rangle$, and $p_{i,\mathbf{k}} = \langle d_{i,-\mathbf{k}} c_{\mathbf{k}} \rangle$, where $c^\dagger_{\mathbf{k}}$ and $d^\dagger_{i,\mathbf{k}}$ ($c_{\mathbf{k}}$ and $d_{i,\mathbf{k}}$) denote the creation (annihilation) of electrons and holes with $i, j = h, l$ refering to HH and LH, respectively. The equations of motion for these variables are given by

$$\frac{d}{dt} f^e_{\mathbf{k}} = \frac{1}{i\hbar} \sum_i (\mathcal{U}_{i,\mathbf{k}} p^*_{i,\mathbf{k}} - \mathcal{U}^*_{i,\mathbf{k}} p_{i,\mathbf{k}}) + \frac{d}{dt} f^e_{\mathbf{k}} \Big|^{col} , \tag{1}$$

$$\begin{aligned} \frac{d}{dt} f^h_{ij,-\mathbf{k}} &= \frac{1}{i\hbar} \sum_l (\mathcal{E}^h_{jl,-\mathbf{k}} f^h_{il,-\mathbf{k}} - \mathcal{E}^h_{li,-\mathbf{k}} f^h_{lj,-\mathbf{k}}) \\ &\quad + \frac{1}{i\hbar} (\mathcal{U}_{j,\mathbf{k}} p^*_{i,\mathbf{k}} - \mathcal{U}^*_{i,\mathbf{k}} p_{j,\mathbf{k}}) + \frac{d}{dt} f^h_{ij,-\mathbf{k}} \Big|^{col} , \end{aligned} \tag{2}$$

$$\begin{aligned} \frac{d}{dt} p_{i,\mathbf{k}} &= \frac{1}{i\hbar} \sum_j (\mathcal{E}^e_{\mathbf{k}} \delta_{ij} + \mathcal{E}^h_{ij,-\mathbf{k}}) p_{j,\mathbf{k}} \\ &\quad + \frac{1}{i\hbar} \sum_j \mathcal{U}_{j,\mathbf{k}} (\delta_{ij} - \delta_{ij} f^e_{\mathbf{k}} - f^h_{ji,-\mathbf{k}}) + \frac{d}{dt} p_{i,\mathbf{k}} \Big|^{col} . \end{aligned} \tag{3}$$

Here, the Hartree-Fock renormalized energies

$$\mathcal{E}^e_{\mathbf{k}} = \epsilon^e_{\mathbf{k}} - \sum_{\mathbf{q}} V_{\mathbf{q}} f^e_{\mathbf{k}+\mathbf{q}} , \tag{4}$$

$$\mathcal{E}^h_{ij,\mathbf{k}} \;=\; \epsilon^h_{i,\mathbf{k}}\delta_{ij} - \sum_{\mathbf{q}} V_{\mathbf{q}} f^h_{ji,\mathbf{k}+\mathbf{q}} \ , \tag{5}$$

$\epsilon^e_{\mathbf{k}}$ and $\epsilon^h_{i,\mathbf{k}}$ denoting the single particle energies of electrons and holes and $V_{\mathbf{q}}$ being the Coulomb matrix element, include the effect of band gap renormalization, and the Hartree-Fock renormalized field

$$\mathcal{U}_{i,\mathbf{k}} = -M_{i,\mathbf{k}}E_0(t)e^{-i\omega_L t} - \sum_{\mathbf{q}} V_{\mathbf{q}} p_{i,\mathbf{k}+\mathbf{q}} \ , \tag{6}$$

$M_{i,\mathbf{k}}$ being the dipole matrix element and $E_0(t)$ (ω_L) denoting the amplitude (central frequency) of the driving field, give rise to excitonic effects and Coulomb enhancement. In these equations only Coulomb terms conserving the number of carriers in each band have been taken into account which are the dominant terms due to their small-q behavior. Thus, contributions between conduction and valence bands leading, e.g., to Auger recombination and impact ionization as well as contributions leading to transitions between the valence bands are neglected. The resulting SBE are equivalent to the multisubband case given in [10].

The last term on the right hand side in each of the Eqs. (1)-(3) denotes the collision term. In the present section dephasing is treated in terms of simple relaxation time T_2 while relaxation processes of the distribution function are neglected. A detailed quantum kinetic treatment of the collsion terms due to carrier-phonon interaction will be discussed in the following sections.

For the case of excitation mainly in the excitonic region of the spectrum the SBE give more insight into the coherent dynamics after transformation into the exciton basis.[10, 11] The frequency ω_n and wave function $p^n_{i,\mathbf{k}}$ of the exciton state $|n\rangle$ in k-space representation are obtained by inserting the ansatz

$$p_{i,\mathbf{k}}(t) = p^n_{i,\mathbf{k}} e^{-i\omega_n t} \tag{7}$$

into the homogeneous part of Eq. (3), i.e., from the eigenvalue problem

$$\left(\epsilon^e_{\mathbf{k}} + \epsilon^h_{i,-\mathbf{k}} - \hbar\omega_n\right) p^n_{i,\mathbf{k}} - \sum_{\mathbf{q}} V_{\mathbf{q}} p^n_{i,\mathbf{k}+\mathbf{q}} = 0 \ . \tag{8}$$

Then, the polarization can be expanded in the complete and orthogonal basis set of exciton states according to

$$p_{i,\mathbf{k}}(t) = \sum_n P_n(t) p^n_{i,\mathbf{k}} \tag{9}$$

where P_n denotes the amplitude of the n-th exciton state. After some calculations the SBE in the exciton basis are obtained:

$$\frac{d}{dt} f^e_{\mathbf{k}} \;=\; -\frac{1}{i\hbar}\sum_{i,n}\left(p^{n*}_{i,\mathbf{k}} M_{i,\mathbf{k}} E_0(t) e^{-i\omega_L t} P^*_n - p^n_{i,\mathbf{k}} M^*_{i,\mathbf{k}} E^*_0(t) e^{i\omega_L t} P_n\right)$$

$$- \frac{1}{i\hbar} \sum_{nn'} \sum_{i,\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'} \left(p^{n*}_{i,\mathbf{k}} p^{n'}_{i,\mathbf{k}'} - p^{n*}_{i,\mathbf{k}'} p^{n'}_{i,\mathbf{k}} \right) P^*_n P_{n'} \ , \qquad (10)$$

$$\frac{d}{dt} f^h_{ij,-\mathbf{k}} = \frac{1}{i\hbar} \sum_l (\mathcal{E}^h_{jl,-\mathbf{k}} f^h_{il,-\mathbf{k}} - \mathcal{E}^h_{li,-\mathbf{k}} f^h_{lj,-\mathbf{k}})$$
$$- \frac{1}{i\hbar} \sum_n \left(p^{n*}_{i,\mathbf{k}} M_{j,\mathbf{k}} E_0(t) e^{-i\omega_L t} P^*_n - p^n_{j,\mathbf{k}} M^*_{i,\mathbf{k}} E^*_0(t) e^{i\omega_L t} P_n \right)$$
$$- \frac{1}{i\hbar} \sum_{nn'} \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'} \left(p^{n*}_{i,\mathbf{k}} p^{n'}_{j,\mathbf{k}'} - p^{n*}_{i,\mathbf{k}'} p^{n'}_{j,\mathbf{k}} \right) P^*_n P_{n'} \ , \qquad (11)$$

$$\frac{d}{dt} P_n = -i\omega_n P_n - \frac{1}{i\hbar} \mu_n E_0(t) e^{-i\omega_L t} - \frac{P_n}{T_2}$$
$$+ \frac{1}{i\hbar} \sum_{ij,\mathbf{k}} p^{n*}_{i,\mathbf{k}} M_{j,\mathbf{k}} E_0(t) e^{-i\omega_L t} \left(\delta_{ij} f^e_{\mathbf{k}} + f^h_{ji,-\mathbf{k}} \right) + \frac{1}{i\hbar} \sum_{n'} P_{n'}$$
$$\times \sum_{\substack{ij \\ \mathbf{k}\mathbf{k}'}} V_{\mathbf{k}-\mathbf{k}'} \left(\delta_{ij} f^e_{\mathbf{k}} + f^h_{ji,-\mathbf{k}} \right) \left(p^{n'*}_{i,\mathbf{k}} p^{n'}_{j,\mathbf{k}'} - p^{n*}_{i,\mathbf{k}'} p^{n'}_{j,\mathbf{k}'} \right) . \qquad (12)$$

The coupling to the light field is now determined by the exciton dipole moment

$$\mu_n = \sum_{i,\mathbf{k}} p^{n*}_{i,\mathbf{k}} M_{i,\mathbf{k}}. \qquad (13)$$

The various terms in the equations of motion can be classified according to the order of the light field: The dominant contribution to the exciton amplitude P_n (first line in Eq. (12)) is of first order in the field. Thus, all driving terms in Eqs. (10),(11) are of second order in the field. The Coulomb terms cannot be neglected in the calculation of the carrier distribution even at low densities. However, these terms cancel in the equation of motion for the complete carrier (or exciton) density

$$N = n^e = n^h = \sum_{\mathbf{k}} f^e_{\mathbf{k}} = \sum_{i,\mathbf{k}} f^h_{ii,\mathbf{k}} \qquad (14)$$

which, since $V_{\mathbf{k}-\mathbf{k}'} = V_{\mathbf{k}'-\mathbf{k}}$, is given by

$$\frac{d}{dt} N = -\frac{1}{i\hbar} \sum_n \left(\mu_n E_0(t) e^{-i\omega_L t} P^*_n - \mu^*_n E^*_0(t) e^{i\omega_L t} P_n \right) . \qquad (15)$$

The second and third line in Eq. (12), which originate from phase space filling, band gap and renormalization, involve products of field or exciton amplitudes and distribution functions. Therefore they are at least of third order in the field. If screening is taken into account additional contributions appear due to the density-dependence of the Coulomb matrix elements which, again, are at least of third order in the field. Thus, in the low density limit, i.e., up to second order in the field, the SBE in the exciton basis (Eqs. (12),(15)) agree with the optical Bloch equations for an ensemble of two-level systems with frequencies ω_n, N denoting the total occupation of the upper states. In this representation the linear response as well as the exciton density in second order are exactly determined by optical Bloch equations which enables one to interpret the results in terms of rotations of a Bloch vector [3, 5]. If higher order terms are included, this analogy does not hold anymore since the full k-dependence of the distribution functions is required for the third-order contributions.

In a coherent control experiment, HL beats can be observed in the exciton density as a function of the pulse delay [4] and in the four wave mixing (FWM) signal [3]. They also modulate pump-probe signals like the differential transmission and reflection. In a coherent control FWM experiment two pulses in the direction $\mathbf{k}_1$ generate a polarization in the sample. A third pulse in the direction $\mathbf{k}_2$ interacts with this polarization and is self-diffracted in the direction $2\mathbf{k}_2 - \mathbf{k}_1$ where the FWM signal is measured. In a first approximation the FWM signal is a measure for the polarization induced by the first two pulses and, since the inhomogeneous broadening is eliminated by a time-reversal, the incoherently summed polarization $\sum_{j,\mathbf{k}} |p_{j,\mathbf{k}}|$ is a good measure for the signal in the sense that it contains the essential features. A full calculation of FWM signals is also possible, in the present case, however, calculations including three pulses for a large number of time delays are required which would be very time consuming.

Figure 1 shows the incoherently summed polarization for two series of time delays between the pulses one and two (150 fs pulses centered at the HH exciton), n refering to the number of HH exciton periods $T_{hh} = 2\pi/\omega_{hh}$ and n_l refering to the respective number of LH periods. For an integer value of n the polarization is enhanced by the second pulse (constructive interference), for an integer plus one half value it is reduced (destructive interference). Due to the dephasing ($T_2 = 2.5$ ps) it is not completely destroyed. The quantum beats, on the other hand, are determined by n_l because the light holes are excited much weaker. The beats vanish completely if n_l assumes an integer plus one half value since in this case the LH component is essentially destroyed by the second pulse.

3. Carrier-phonon quantum kinetics: Phonon quantum beats

On the semiclassical kinetic level interaction mechanisms lead to transitions between states, the transtition rates being obtained from Fermi's golden rule. On this level, these mechanisms are a source of dephasing of coherent variables like interband, intersubband, and intervalence band

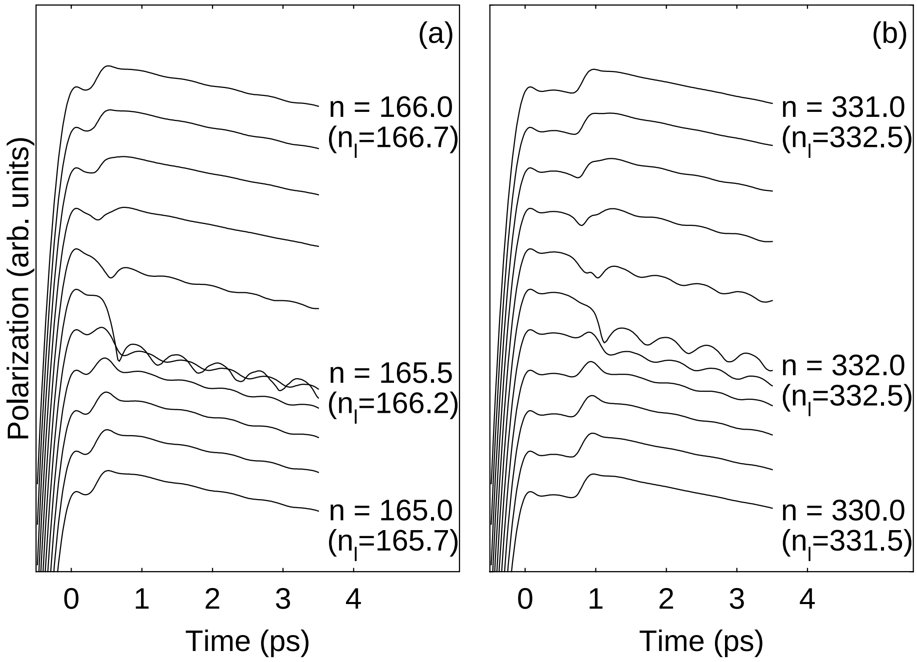

Figure 1. Coherent control of HL quantum beats in a single quantum well. The time delay between the pulses is given in units of the HH period (n) and the LH period (n_l).

polarizations. On a quantum mechanical level, on the other hand, they can also be a source of coherence due to the coupling between states. In the case of carrier-LO-phonon interaction it has been shown theoretically [12, 13, 14, 15] and experimentally [16] that in a quantum kinetic treatment the decay of the polarization exhibits an oscillatory contribution, the phonon quantum beats. Only recently it has been shown experimentally that also these beats can be coherently controlled [8, 9].

Carrier-phonon quantum kinetics in a two-band model can be described on various levels, in particular nonequilibrium Green's functions [12, 13] or the density matrix formalism [14, 15], both leading to the same equations of motion on certain approximation levels. The carrier-phonon Hamiltonian introduces new variables in the eqations of motion of the single particle density matrices $f^{e,h}_{\mathbf{k}}$, $p_{\mathbf{k}}$ and $n_{\mathbf{q}} = \langle b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \rangle$, these are the phonon assisted density matrices

$$S^e_{\mathbf{k+q,k}} = \frac{i}{\hbar} g_{\mathbf{q}} \langle c^\dagger_{\mathbf{k+q}} b_{\mathbf{q}} c_{\mathbf{k}} \rangle \ , \quad S^h_{\mathbf{k+q,k}} = -\frac{i}{\hbar} g_{\mathbf{q}} \langle d^\dagger_{\mathbf{k+q}} b_{\mathbf{q}} d_{\mathbf{k}} \rangle \ , \qquad (16)$$

$$T^{(+)}_{\mathbf{k+q,k}} = \frac{i}{\hbar} g_{\mathbf{q}} \langle d_{-(\mathbf{k+q})} b_{\mathbf{q}} c_{\mathbf{k}} \rangle \ , \quad T^{(-)}_{\mathbf{k,k+q}} = \frac{i}{\hbar} g_{\mathbf{q}} \langle d_{-\mathbf{k}} b^\dagger_{\mathbf{q}} c_{\mathbf{k+q}} \rangle \ , \qquad (17)$$

$g_{\mathbf{q}}$ being the coupling matrix elements and $b^\dagger_{\mathbf{q}}$ ($b_{\mathbf{q}}$) denoting phonon creation (annihilation) operators. They constitute the starting point of an infinite hierarchy of equations of motion involving higher order density matrices. In terms of a correlation expansion, this hierarchy is truncated by factorization on a certain level. A discussion of this factorization can be found in [17].

The Hamiltonian for a two-band model including carrier-carrier and carrier-phonon interaction is given by

$$\begin{aligned} H = & \sum_{\mathbf{k}} \left(\epsilon^e_{\mathbf{k}} c^\dagger_{\mathbf{k}} c_{\mathbf{k}} + \epsilon^h_{\mathbf{k}} d^\dagger_{\mathbf{k}} d_{\mathbf{k}} \right) + \sum_{\mathbf{q}} \hbar\omega_{op}\, b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \\ - & \sum_{\mathbf{k}} \left(M_{\mathbf{k}} E_0(t) e^{-i\omega_L t}\, c^\dagger_{\mathbf{k}} d^\dagger_{-\mathbf{k}} + M^*_{\mathbf{k}} E^*_0(t) e^{i\omega_L t}\, d_{-\mathbf{k}} c_{\mathbf{k}} \right) \\ + & \sum_{\mathbf{k},\mathbf{q}} g_{\mathbf{q}} \left(c^\dagger_{\mathbf{k}+\mathbf{q}} b_{\mathbf{q}} c_{\mathbf{k}} - c^\dagger_{\mathbf{k}} b^\dagger_{\mathbf{q}} c_{\mathbf{k}+\mathbf{q}} - d^\dagger_{\mathbf{k}+\mathbf{q}} b_{\mathbf{q}} d_{\mathbf{k}} + d^\dagger_{\mathbf{k}} b^\dagger_{\mathbf{q}} d_{\mathbf{k}+\mathbf{q}} \right) \\ + & \sum_{\substack{\mathbf{k},\mathbf{k}' \\ \mathbf{q}}} \frac{V_{\mathbf{q}}}{2} \left(c^\dagger_{\mathbf{k}} c^\dagger_{\mathbf{k}'} c_{\mathbf{k}'+\mathbf{q}} c_{\mathbf{k}-\mathbf{q}} + d^\dagger_{\mathbf{k}} d^\dagger_{\mathbf{k}'} d_{\mathbf{k}'+\mathbf{q}} d_{\mathbf{k}-\mathbf{q}} - 2 c^\dagger_{\mathbf{k}} d^\dagger_{\mathbf{k}'} d_{\mathbf{k}'+\mathbf{q}} c_{\mathbf{k}-\mathbf{q}} \right) . \end{aligned} \tag{18}$$

If all four-point correlations are neglected, this Hamiltonian gives rise to the equations of motion for the single particle density matrices

$$\begin{aligned} \frac{d}{dt} f^e_{\mathbf{k}} &= 2\mathrm{Re}\left\{ \frac{i}{\hbar} \mathcal{U}^*_{\mathbf{k}} p_{\mathbf{k}} \right\} \\ &+ \sum_{\mathbf{q}} \left[2\mathrm{Re}\left\{ S^e_{\mathbf{k}+\mathbf{q},\mathbf{k}} \right\} - 2\mathrm{Re}\left\{ S^e_{\mathbf{k},\mathbf{k}-\mathbf{q}} \right\} \right] , \end{aligned} \tag{19}$$

$$\begin{aligned} \frac{d}{dt} f^h_{-\mathbf{k}} &= 2\mathrm{Re}\left\{ \frac{i}{\hbar} \mathcal{U}^*_{\mathbf{k}} p_{\mathbf{k}} \right\} \\ &+ \sum_{\mathbf{q}} \left[2\mathrm{Re}\left\{ S^h_{-(\mathbf{k}+\mathbf{q}),-\mathbf{k}} \right\} - 2\mathrm{Re}\left\{ S^h_{-\mathbf{k},-(\mathbf{k}-\mathbf{q})} \right\} \right] , \end{aligned} \tag{20}$$

$$\begin{aligned} \frac{d}{dt} p_{\mathbf{k}} &= -\frac{i}{\hbar} \left(\mathcal{E}^e_{\mathbf{k}} + \mathcal{E}^h_{-\mathbf{k}} \right) p_{\mathbf{k}} - \frac{i}{\hbar} \mathcal{U}_{\mathbf{k}} \left(1 - f^e_{\mathbf{k}} - f^h_{-\mathbf{k}} \right) \\ &+ \sum_{\mathbf{q}} \left[T^{(+)}_{\mathbf{k}+\mathbf{q},\mathbf{k}} - T^{(-)}_{\mathbf{k}-\mathbf{q},\mathbf{k}} - T^{(+)}_{\mathbf{k},\mathbf{k}-\mathbf{q}} + T^{(-)}_{\mathbf{k},\mathbf{k}+\mathbf{q}} \right] , \end{aligned} \tag{21}$$

$$\frac{d}{dt} n_{\mathbf{q}} = \sum_{\mathbf{k}} \left[2\mathrm{Re}\left\{ S^e_{\mathbf{k}+\mathbf{q},\mathbf{k}} \right\} + 2\mathrm{Re}\left\{ S^h_{\mathbf{k}+\mathbf{q},\mathbf{k}} \right\} \right] . \tag{22}$$

For the phonon assisted density matrix $T^{(+)}$ we obtain

$$\begin{aligned} \frac{d}{dt} T^{(+)}_{\mathbf{k}',\mathbf{k}} &= \frac{1}{i\hbar} \left(\mathcal{E}^h_{-\mathbf{k}'} + \mathcal{E}^e_{\mathbf{k}} + \hbar\omega_{op} \right) T^{(+)}_{\mathbf{k}',\mathbf{k}} - \frac{i}{\hbar} \left(1 - f^h_{-\mathbf{k}'} - f^e_{\mathbf{k}} \right) \tilde{T}^{(+)}_{\mathbf{k}',\mathbf{k}} \\ &+ \frac{i}{\hbar} \left(\mathcal{U}_{\mathbf{k}'} S^e_{\mathbf{k}',\mathbf{k}} - \mathcal{U}_{\mathbf{k}} S^h_{-\mathbf{k},-\mathbf{k}'} \right) - \frac{i}{\hbar} \left(p_{\mathbf{k}'} \tilde{S}^e_{\mathbf{k}',\mathbf{k}} - p_{\mathbf{k}} \tilde{S}^h_{-\mathbf{k},-\mathbf{k}'} \right) \end{aligned}$$

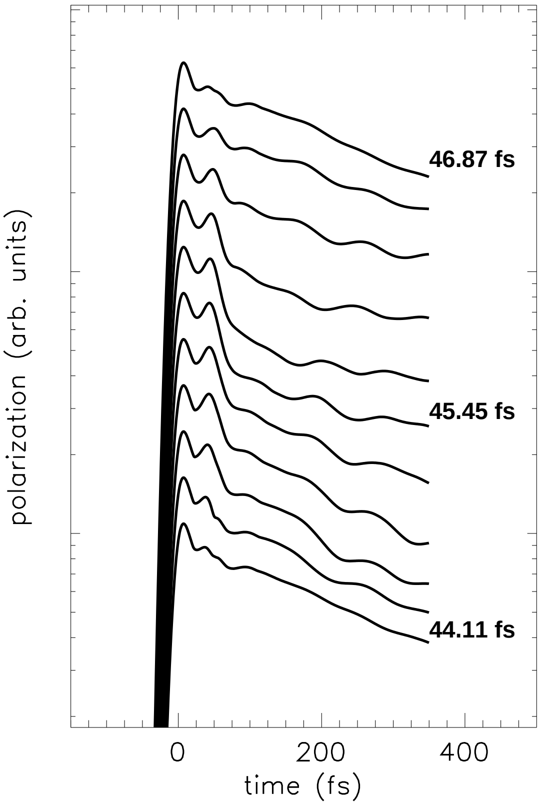

Figure 2. Coherent control of phonon quantum beats in a quantum wire for the case of excitation by two 15 fs pulses centered at the exciton frequency.

$$
\begin{aligned}
&+ \frac{1}{\hbar^2}|g_{\mathbf{k}'-\mathbf{k}}|^2 \left[\left(n_{\mathbf{k}'-\mathbf{k}}+1\right)\left(1-f^e_{\mathbf{k}}\right)+n_{\mathbf{k}'-\mathbf{k}}f^e_{\mathbf{k}}\right] p_{\mathbf{k}'} \\
&- \frac{1}{\hbar^2}|g_{\mathbf{k}'-\mathbf{k}}|^2 \left[\left(n_{\mathbf{k}'-\mathbf{k}}+1\right)\left(1-f^h_{-\mathbf{k}'}\right)+n_{\mathbf{k}'-\mathbf{k}}f^h_{-\mathbf{k}'}\right] p_{\mathbf{k}} \qquad (23)
\end{aligned}
$$

with the off-diagonal Hartree-Fock terms

$$\tilde{S}^{e,h}_{\mathbf{k}',\mathbf{k}} = -\sum_{\mathbf{q}'} V_{\mathbf{q}'} S^{e,h}_{\mathbf{k}'+\mathbf{q}',\mathbf{k}+\mathbf{q}'} + V_{\mathbf{k}'-\mathbf{k}} \sum_{\mathbf{q}'} \left(S^{e,h}_{\mathbf{k}'+\mathbf{q}',\mathbf{k}+\mathbf{q}'} + S^{h,e}_{\mathbf{k}'+\mathbf{q}',\mathbf{k}+\mathbf{q}'} \right) \qquad (24)$$

$$\tilde{T}^{(\pm)}_{\mathbf{k}',\mathbf{k}} = -\sum_{\mathbf{q}'} V_{\mathbf{q}'} T^{(\pm)}_{\mathbf{k}'+\mathbf{q}',\mathbf{k}+\mathbf{q}'}, \qquad (25)$$

and similar equations for the other phonon assisted density matrices.

Figure 2 shows the incoherently summed polarization for the case of a quantum wire excited by two 15 fs pulses centered at the exciton frequency.

The oscillations, which in the present case are due to a beating between the exciton and a phonon sideband above the exciton, can be clearly controlled by the relative phase of the second pulse. The beats are essentially absent for the lowest and the highest value of the time delay shown in the figure. These values correspond to 16.5 and 17.5 times the period of the phonon sideband which is determined by the sum of exciton and beat frequency. Thus, the behavior is the same as in the case of HL beats shown above. It is interesting to notice that the decay of the signal is modified by the second pulse, a feature which has been found also in the experiment [8, 9].

4. Non-perturbative treatment of carrier-phonon interaction

The quantum kinetic theory discussed in the previous section is based on a correlation expansion which assumes that correlations between an increasing number of quasiparticles are of decreasing importance. Since this expansion is related to an expansion in the carrier-phonon coupling matrix element, the analysis of systems with a stronger phonon coupling requires to take into account higher orders in the hierarchy. In this section we discuss an alternative method which has been proposed recently [18] and which avoids the hierarchy. This method will be applied to a simple model of a two-level system interacting with a single phonon mode. This model, which has already been shown to explain very well the coherent control of phonon quantum beats in the weak coupling limit [8, 9], can be solved exactly by the present method.

The basic idea is to substitute the single particle density matrices by generating functions defined according to, e.g.,

$$f^e_{\mathbf{k}\mathbf{k}'}(\{\alpha_\mathbf{q}\},\{\beta_\mathbf{q}\}) = \langle c^\dagger_\mathbf{k} c_{\mathbf{k}'} \exp[\sum_\mathbf{q} \alpha_\mathbf{q} b^\dagger_\mathbf{q}] \exp[\sum_\mathbf{q} \beta_\mathbf{q} b_\mathbf{q}]\rangle \,, \tag{26}$$

$$p_{\mathbf{k}\mathbf{k}'}(\{\alpha_\mathbf{q}\},\{\beta_\mathbf{q}\}) = \langle d_\mathbf{k} c_{\mathbf{k}'} \exp[\sum_\mathbf{q} \alpha_\mathbf{q} b^\dagger_\mathbf{q}] \exp[\sum_\mathbf{q} \beta_\mathbf{q} b_\mathbf{q}]\rangle \tag{27}$$

with complex numbers $\alpha_\mathbf{q}$ and $\beta_\mathbf{q}$. All phonon-related higher order density matrices can be obtained as partial derivatives with respect to $\alpha_\mathbf{q}$ and $\beta_\mathbf{q}$ taken at $\alpha_\mathbf{q} = \beta_\mathbf{q} = 0$. As a result, the hierarchy of equations of motion is transformed into a set of partial differential equations. While the treatment of a full semiconductor model is too complicated due to the high dimension of the equations, the following two-level model interacting with one phonon mode, which stands for a coupled exciton-phonon system, can be solved analytically.

The Hamiltonian including the coupling to the light field is given by

$$H = \hbar\Omega c^\dagger c + \hbar\omega_L b^\dagger b + \hbar g\,(b^\dagger + b)\, c^\dagger c - \hbar M_0\,(E\, c^\dagger d^\dagger + E^* dc)\ . \tag{28}$$

The dynamical variables in this case are the functions

$$p(\alpha,\beta,t) = \langle dc\exp(\alpha b^\dagger)\exp(\beta b)\rangle\ , \quad (29)$$
$$f(\alpha,\beta,t) = \langle c^\dagger c\exp(\alpha b^\dagger)\exp(\beta b)\rangle\ , \quad (30)$$
$$n(\alpha,\beta,t) = \langle \exp(\alpha b^\dagger)\exp(\beta b)\rangle\ , \quad (31)$$

which obey the following equations of motion:

$$i\partial_t p = [\Omega + \omega_L(\beta\partial_\beta - \alpha\partial_\alpha) + g(\beta + \partial_\alpha + \partial_\beta)]p - M_0 E(n-2f), \quad (32)$$
$$i\partial_t f = [\omega_L(\beta\partial_\beta - \alpha\partial_\alpha) + g(\beta - \alpha)]f - M_0[Ep^T - E^* p], \quad (33)$$
$$i\partial_t n = \omega_L(\beta\partial_\beta - \alpha\partial_\alpha)n + g(\beta-\alpha)f, \quad (34)$$

with $p^T(\alpha,\beta) \equiv p^*(\beta^*,\alpha^*)$. By means of the transformations

$$\bar{p}(\alpha,\beta,t) = \mathrm{e}^{i\bar{\Omega}t+\beta\gamma\mathrm{e}^{i\omega_L t}}\, p(\alpha\mathrm{e}^{-i\omega_L t}+\gamma,\, \beta\,\mathrm{e}^{i\omega_L t}-\gamma, t)\ , \quad (35)$$
$$\bar{f}(\alpha,\beta,t) = \mathrm{e}^{\gamma(\beta\mathrm{e}^{i\omega_L t}+\alpha\mathrm{e}^{-i\omega_L t})}\, f(\alpha\,\mathrm{e}^{-i\omega_L t},\, \beta\,\mathrm{e}^{i\omega_L t}, t)\ , \quad (36)$$
$$\bar{n}(\alpha,\beta,t) = n(\alpha\,\mathrm{e}^{-i\omega_L t},\, \beta\,\mathrm{e}^{i\omega_L t}, t)\ , \quad (37)$$
$$\bar{E}(t) = M_0\,\exp[i\bar{\Omega}t]E(t), \quad (38)$$

with $\bar{\Omega} = \Omega - \omega_L\gamma^2$, and $\gamma = g/\omega_L$ the partial derivatives are eliminated resulting in a set of three ordinary differential equations which can be further simplified by setting $\bar{y}(\alpha,t) = \bar{y}(-\alpha^*,\alpha,t)$ where y stands for p, f, and n. The final equations of motion are

$$\partial_t\bar{p}(\alpha) = i\bar{E}\,[\ \mathrm{e}^{\alpha\gamma\,\mathrm{e}^{i\omega_L t}}\ \bar{n}(\alpha-\gamma\mathrm{e}^{-i\omega_L t}) - 2\,\mathrm{e}^{\alpha^*\gamma\,\mathrm{e}^{-i\omega_L t}}\ \bar{f}(\alpha-\gamma\mathrm{e}^{-i\omega_L t})\,], \quad (39)$$

$$\partial_t\bar{f}(\alpha) = i\mathrm{e}^{-\gamma^2}[\ \bar{E}\ \mathrm{e}^{\alpha\gamma\,\mathrm{e}^{i\omega_L t}}\ \bar{p}^*(-\alpha+\gamma\mathrm{e}^{-i\omega_L t}) - \bar{E}^*\,\mathrm{e}^{-\alpha^*\gamma\,\mathrm{e}^{-i\omega_L t}}\ \bar{p}(\alpha+\gamma\mathrm{e}^{-i\omega_L t})\,], \quad (40)$$

$$\partial_t\bar{n}(\alpha) = -i\gamma\,\omega_L\,2\,\mathrm{Re}[\alpha e^{i\omega_L t}]\,e^{-\gamma\,2i\mathrm{Imm}[\alpha\,\mathrm{e}^{i\omega_L t}]}\ \bar{f}(\alpha). \quad (41)$$

These equations have to be solved with the initial conditions $\bar{f}(\alpha,0) = \bar{p}(\alpha,0) = 0$ and $\bar{n}(\alpha,0) = \exp[-n_L|\alpha|^2]$, n_L being the equilibrium (Bose) value of the phonon occupation. From the dynamical variables we obtain the polarization $P = \hbar M_0\mathrm{e}^{-(i\bar{\Omega}t+\gamma^2)}\bar{p}(\gamma\,\mathrm{e}^{-i\omega_L t},t)$ and the exciton density $N_e = \bar{f}(\alpha=0,t)$.

The equations can be solved iteratively for the case of excitation by δ-pulses $E(t) = E_0\,\delta(t)$. The linear polarization is given by

$$P_\delta^{(1)}(t) = i\hbar M_0^2 E_0\,\theta(t)\,\mathrm{e}^{-i\bar{\Omega}t+\gamma^2\,(\mathrm{e}^{-i\omega_L t}-1-n_L\,|\mathrm{e}^{-i\omega_L t}-1|^2)}\ . \quad (42)$$

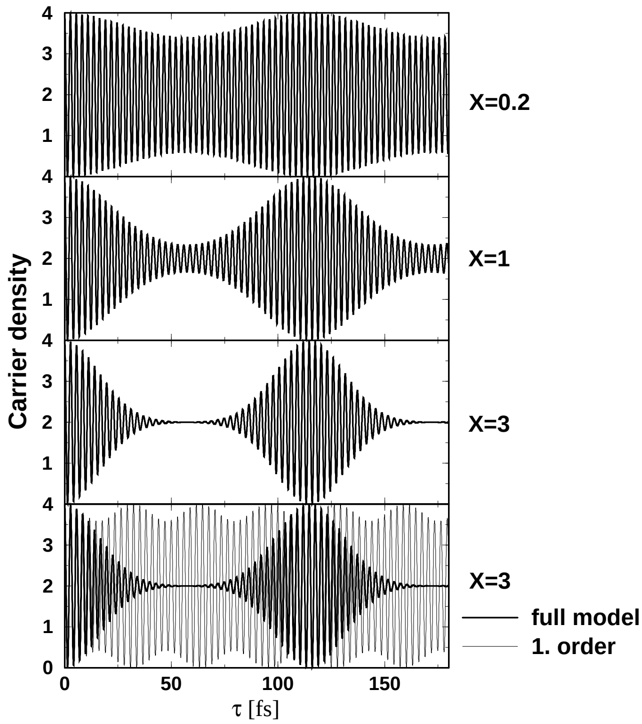

Figure 3. Coherent control of carrier density obtained from the two-level model for 15 fs pulses at T = 10K.

At zero temperature, where $n_L = 0$, this result can also be written as

$$P_{\delta}^{(1)}(t) = i\hbar M_0^2 E_0 \theta(t) \sum_{j=0}^{\infty} \mathrm{e}^{-\gamma^2} \frac{\gamma^{2j}}{j!} \mathrm{e}^{-i(\bar{\Omega}+j\omega_L)t} \tag{43}$$

which shows the well-known result of phonon sidebands at multiples of the phonon frequency above the polaron shifted exciton energy. It is interesting to compare this exact result with the result obtained from the first order correlation expansion:

$$P_{\delta corr}^{(1)}(t) = \frac{i\hbar M_0^2 E_0 \theta(t)}{(1+2x)} \{(x+1)\mathrm{e}^{-i[\Omega-\omega_L x]t} + x\mathrm{e}^{-i[\Omega+\omega_L(1+x)]t}\} \tag{44}$$

with $x = (\sqrt{1+4\gamma^2}-1)/2 \approx \gamma^2$. As expected, we find only one phonon sideband, the frequency however being shifted by a factor $(1+2x)$.

The exciton density up to $\mathcal{O}(E^2)$ generated by two δ-pulses with a time delay τ is given by

$$N_{e,\delta}^{(2)}(t,\tau) \quad = \quad |M_0 E_0|^2 \; \{\theta(t) + \theta(t-\tau)$$

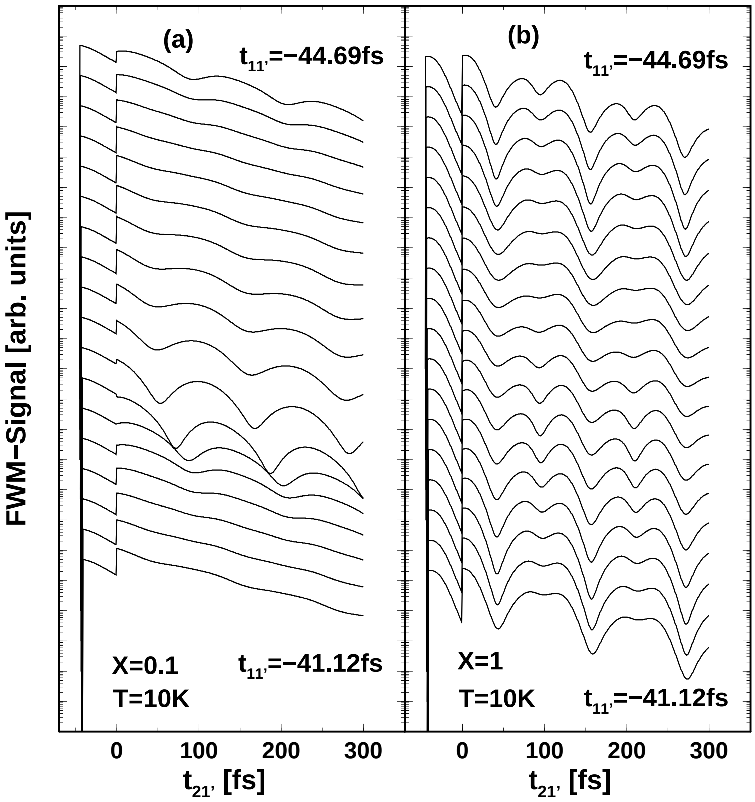

Figure 4. Coherent control of phonon quantum beats obtained from the two-level model for delta pulses at T = 10K.

$$\times[2\mathrm{Re}[\mathrm{e}^{-i\bar{\Omega}\tau+\gamma^2\,(\mathrm{e}^{-i\omega_L\tau}-1-n_L\,|\mathrm{e}^{-i\omega_L\tau}-1|^2)}]+1]\} \quad (45)$$

exhibiting the fast coherent control oscillations at the polaron shifted exciton energy $\bar{\Omega}$ as well as additional structures related to the phonon coupling. In Fig. 3 the exciton density created by two 15 fs pulses at a temperature of 10 K is shown as a function of τ for three different values of the phonon coupling. For $x = 0.2$ we find a weak modulation with the phonon frequency. With increasing coupling the higher order terms do not show up as additional frequencies, instead the minima become more pronounced leading finally to a behavior similar to a “collapse and revival”. In the lowest part of the figure we compare the full model with the first order correlation expansion result which, of course, are quite different at this high value of x. Nevertheless it is interesting to notice that the very initial part below 20 fs is still in agreement which is consistent with the interpretation that the correlations are successively built up.

For the case of excitation by three δ-pulses the FWM signal can be calculated analytically. The polarization in the FWM direction is given by

$$\begin{aligned} P^{(3)}_{2k_2-k_1} &= -i\hbar M_0^2 E_0\,|M_0E_0|^2\theta(t)\mathrm{e}^{-i\bar{\Omega}t+2\gamma^2(\cos(\omega_L t)-1)} \\ &\times\{\theta(-t_1)\mathrm{e}^{-i\bar{\Omega}t_1+\gamma^2(2\mathrm{e}^{-i\omega_L t_1}-\mathrm{e}^{i\omega_L(t-t_1)}-1-n_L|2-\mathrm{e}^{-i\omega_L t_1}-\mathrm{e}^{-i\omega_L t}|^2)} \\ &+\theta(-t_{1'})\mathrm{e}^{-i\bar{\Omega}t_{1'}+\gamma^2(2\mathrm{e}^{-i\omega_L t_{1'}}-\mathrm{e}^{i\omega_L(t-t_{1'})}-1-n_L|2-\mathrm{e}^{-i\omega_L t_{1'}}-\mathrm{e}^{-i\omega_L t}|^2)}\} \end{aligned} \tag{46}$$

where the time of the pulse in direction $\mathbf{k}_2$ has been set to zero. The FWM signal $P^{(3)}_{2k_2-k_1}$ for different time delays of the pulses in direction $\mathbf{k}_1$ and for two values of the coupling constants are shown in Fig. 4. The results for the weak coupling are in agreement with those given in [8] and clearly show the coherent control of the quantum beats. By an expansion of the exponential (for $t=0$, $n_L=0$, $t_{21}>0$, $t_{21'}>0$) the signal can be written as

$$|P^{(3)}_{2k_2-k_1}|^2 = const\,\{1+\cos(\bar{\Omega}t_{11'})+\gamma^2\,h(t_{21'},t_{11'})\} \tag{47}$$

with $h(t_{21'},t_{11'}) = \cos\omega_L t_{21'}\{\cos\omega_L t_{11'}+1+\cos(\bar{\Omega}+\omega_L)t_{11'}+\cos\bar{\Omega}t_{11'}\}$ $+\sin\omega_L t_{21'}\{\sin\omega_L t_{11'}+\sin(\bar{\Omega}+\omega_L)t_{11'}-\sin\bar{\Omega}t_{11'}\}$ which directly shows that the beat amplitude h vanishes for $(\bar{\Omega}+\omega_L)t_{11'}=(2n+1)\pi$ or $\bar{\Omega}t_{11'}=(2n+1)\pi$ in agreement with the results of the previous sections. In the case of a stronger phonon coupling the FWM signal in Fig. 4(b) exhibits higher harmonics of the phonon frequency. Since now more than two transitions are beating the oscillations cannot be anymore switched off completely.

5. Conclusions

We have presented a theoretical analysis of the coherent control of heavy-light hole and phonon quantum beats. In all cases the quantum beats are switched off by the second pulse if there is destructive interference for the weaker of the two beating transitions, which in the first case was the light hole and in the second case the phonon sideband. We have shown by an analytical solution of a two-level model coupled to a single phonon mode that for stronger phonon coupling higher harmonics appear in the FWM signal and a full control of the beats is not anymore possible.

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