Simulating heterodyne four-wave mixing signals of a single quantum dot coupled to a discrete phonon mode

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Nonlinear optical spectroscopy is a powerful technique to investigate the dynamics of charge carriers in semiconductor nanostructures. By using a heterodyne interferometry technique one is able to detect coherence as well as occupation dynamics of a single quantum dot (QD) in a four-wave-mixing (FWM) experiment [1].

Usually the FWM experiment consists of two pulses propagating along two different wavevectors $\mathbf{k}_1$ and $\mathbf{k}_2$ which excite a sample. Pulse 1 is time-shifted to have a delay with respect to pulse 2. The FWM signal can then be extracted along $2\mathbf{k}_2 - \mathbf{k}_1$ in an angle-resolved experiment. The polarization dynamics can be probed by scanning the delay between the pulses. In the heterodyne setup a collinear configuration is used. The exciting fields are not labeled in $\mathbf{k}$-space, but in frequency space resulting in a phase shift of the exciting pulses with respect to each other. Hence, a phase relation of the generated signal can be detected by optical heterodyning. While the angle-resolved FWM relies on spatial homogeneity of the sample, the co-linear excitation allows to measure FWM signals of single QDs. In typical experiments thousands of pulse pairs are used to accumulate the FWM signal.

We model the behavior of a QD in terms of a two-level system, excited by a series of ultrafast pulses. The model is extended by the coupling to a discrete phonon mode in the independent boson model. Even including the phonon coupling for ultrafast optical excitations the dynamics of the entire system can be calculated without any approximations by introducing characteristic functions [2]. Simulating the dynamics of the system, i.e., calculating the characteristic functions during a series of thousands of pulse pairs, it is possible to simulate FWM signals in direct analogy to the experimental realization.

The measured FWM signals of some QDs exhibit photon echos, because of spectral wandering of their transition energies, due for example to charge fluctuations [3]. This effect can be treated theoretically in terms of an ensemble average, which naturally results in the formation of a photon echo. Here, I will show that in our approach the spectral wandering can be treated in the time domain by exciting the system subsequently with many pulses introducing and considering a randomized transition energy. This creates a photon echo in the FWM dynamics. In the FWM-spectrum it appears as an inhomogeneous broadening which can be separated from homogeneous broadening by performing two-dimensional Fourier transform spectroscopy [4]. In a second step I will take the coupling to a discrete phonon mode into account. I will discuss its influence on the exciton dynamics and its impact on FWM signals. In our approach we have also access to the corresponding phonon quantum state which can be visualized in terms of the Wigner function. This can help to interprete the interplay between phonon and exciton dynamics.

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