Coherent contributions to population dynamics in a semiconductor microcavity

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Abstract: Ultrafast exciton-polariton dynamics are investigated using optical multidimensional coherent spectroscopy. A Bloch equations simulation including Pauli blocking and the Coulomb interaction between excitons is found to give good agreement with measured spectra. © 2022 The Author(s)

Microcavities photonically enhance the already relatively strong nonlinear optics of semiconductor nanostructures, enabling novel coherent phenomena and potential applications in quantum information processing. Much about the nonlinear optics of semiconductor nanostructures has been learned by applying optical multidimensional coherent spectroscopy techniques, but only recently have they been applied to nanostructures in microcavities [1-4]. Here we investigate the dependence of rephasing 2D spectra on population time and compare with a theoretical model based on the Bloch equations with the Coulomb interaction treated on a Hartree-Fock level.

The sample is a monolithic, planar semiconductor microcavity grown by molecular beam epitaxy on a GaAs substrate. The structure consists of two GaAs/AlAs distributed Bragg reflectors producing a wedged λ cavity with a single 8-nm, In0.04Ga0.96As quantum well (QW) at its antinode. The structure gives rise to an exciton eigenmode, \( E_X = 1490.1 \text{ meV} \) (832 nm) at 6 K temperature, and a tunable cavity mode, \( E_c(\Delta) \), where the cavity detuning is \( \Delta = E_c - E_X \). Normal-mode splitting near \( \Delta = 0 \) meV mixes the exciton and photon, forming lower polariton and upper polariton (LP and UP) branches that imitate a three-level system in linear spectroscopy.

Measurements are performed with a multidimensional optical nonlinear spectrometer (MONSTR) [5] with 120-fs pulses. Three excitation pulses are incident on the microcavity sample in a box geometry, with the laser wavelength set to equally excite LP and UP polaritons. The beams impinge the sample with incident angle 5°. Four-wave mixing (FWM) emission is collected in the phase-matching direction \( -k_A + k_B + k_c \). Spectral interferograms are recorded with evolution time \( T \) between pulses A and B and population/mixing time \( T \) between pulses B and C. Rephas ing spectra \( S_{\text{SCLH}}(\omega, T, \omega_r) \) are calculated from complex FWM spectra (emitted FWM field as a function of emission frequency \( \omega_r \)) recorded as a function of \( r \). Only co-circular spectra are presented here. This polarization configuration forbids excitation of biexciton-like states through the excitonic selection rules, maintaining a simple two-level scheme.

A selection of rephasing 2D spectra (real part) as a function of \( T \) is shown in Fig. 1(a-d). We observe four strong features throughout the \( T \) dependence - two diagonal intra-action features (LP, UP) and two off-diagonal interaction features (LP-UP and UP-LP). In addition, there are vertical stripes along the \( -\hbar \omega_r \) direction at the emission energies of the LP and UP resonances. Cross-diagonal lineouts as a function of \( T \) are shown in Fig. 1(e-g) and the integrated amplitude of the LP peak is shown as a function of \( T \) in Fig. 1(h). The decay of the intra-action and interaction features show fast and slow components, although with unique decay rates that suggest both energy transfer from UP to LP resonances, with a persistence of UP populations even at longer \( T \).

Numerical simulations of the coupled QW-microcavity system use a set of Bloch equations describing the dynamics of the excitonic transition \( \rho \) driven by the intracavity field \( \alpha \) with incoherent excitation occupation \( \bar{N} \).

\[ \partial_t \alpha = (\gamma_\alpha - i \omega_\alpha) \alpha + igp + \bar{N}E(t), \] (1a)

\[ \partial_t p = (\gamma_p - i \omega_p) p + i g \alpha - (\alpha|p|^2 + \bar{N}) - Vp(|p|^2 + \bar{N}), \] (1b)

\[ \partial_t \bar{N} = -\gamma_N \bar{N} - (\gamma_\alpha - 2\gamma_p) pp^*, \] (1c)

where \( g \) denotes the light-matter coupling strength and \( \bar{N} \) the coupling from the external field \( E(t) \) into the cavity. The exciton resonance frequency is \( \omega_\alpha \), the energetically lowest cavity mode frequency is \( \omega_p \) and \( \gamma_\alpha \) are the exciton dephasing rate and inverse cavity photon lifetime respectively, and \( \gamma_N \) is the incoherent excitation population decay rate. These equations treat the Coulomb-interaction on a Hartree-Fock level. The parameters \( b \) and \( V \) denote...
the optical nonlinearities that give rise to a FWM signal, where \( b \) denotes Pauli blocking and \( V \) the Coulomb interaction between excitons with the same spin. Spectra calculated separately for each contribution to the nonlinearity are shown for \( T = 0.5 \) ps and \( \Delta = 0 \) meV in Fig. 2, along with the full calculation.

![Experimental rephasing spectra at (a) \( T = 0.1 \) ps, (b) 1 ps, (c) 5 ps, and (d) 40 ps. (e-g) Cross-diagonal lineouts of the LP, UP, and LP-UP peaks. (h) The dependence of the LP peak amplitude on \( T \) at three detuning values.](image)

![Theoretical spectra. Pauli blocking and Coulomb interaction contributions and full calculation of the simulated rephasing spectra for \( T = 0.5 \) ps, \( \Delta = 0 \) meV and co-circular polarization. (a)-(e) amplitude and (f)-(j) real parts of the spectra.](image)

We find remarkable agreement between theory and experiment. It should be noted that the model is relatively simple and general, as it simulates a two-level system strongly coupled to the microcavity, and therefore potentially applies to a variety of other material systems. There is only one free parameter for the optical nonlinearity: the ratio between the Pauli blocking and Coulomb interaction terms.

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