Exciton-exciton correlation in the ultrafast nonlinear optical response in the nonperturbative regime

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1. Introduction

The ultrafast nonlinear optical response of semiconductor-microcavities (SMCs) in the nonperturbative regime is attracting growing interest for exploring fundamental light-matter interactions in many-particle quantum systems,[1–3] as well as for the future implementations of ultrafast photonic devices. In spite of many investigations,[4] the coherent dynamics of excitonic nonlinear-response in the nonperturbative regime has not been fully understood. Open questions are how strong exciton-cavity coupling affects exciton-exciton (*X-X*) interactions, and how in turn the *X-X* correlation influences the nonlinear response of SMCs.

We investigate these open questions and show that the nonperturbative regime provides an unprecedented tool for investigating four-particle correlations. By including the full time behaviour of four particle correlations we are able to reproduce and explain the ultrafast nonlinear coherent response which can be observed in pump and probe experiments.[3]

We start from the equations for the third-order exciton polarization and cavity field describing quantum optical effects and coherent nonlinear optics in SMCs,[5] perform the semiclassical factorization and include multiple scattering processes.

For the numerical calculations we consider a GaAs microcavity system with parameters given in the figure caption. We both use a resonant gaussian 100 fs pump pulse 2 with zero delay respect to the probe pulse 1 producing a maximum exciton density $n \approx 0.13\pi a^2$ and also, to discriminate the energy dependence of power dependent absorption and shift, a spectrally narrower pulse with 700fs duration and centered on the lower or upper polariton energy. The probe is a 100fs pulse 10^{-4} weaker of the 100fs pump pulse. In Fig. 1 we display spectra of the calculated time resolved probe reflectivity with (thicker line) and without the presence of the pump beam. Fig. 1(b,c) have been obtained by using the spectrally narrower pump beam centered respectively on the lower and upper polariton energy (shaded spectrum). Fig. 1(a) has been obtained by using the broadband 100fs pump pulse. The agreement with Fig. 2 of Ref. 1 is excellent and impressive.

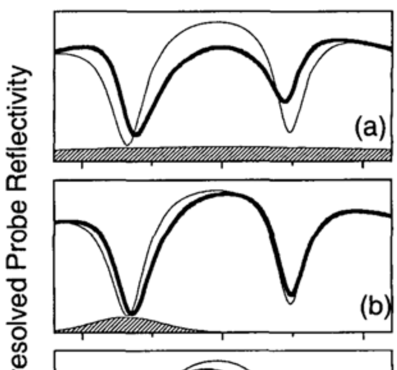

Fig. 1. Spectra of the calculated time-resolved probe reflectivity with the pump switched on (thicker line) and off. (a) broadband pump pulse (shaded spectrum). (b) and (c) Narrowband pump pulses centered on the lower and upper polariton energy. We have used the following parameters for the microcavity system: dipole coupling rate V = 2.85 meV, exciton dephasing rate $\gamma_x = 0.35$ meV, and photon escape rate $\gamma_c = 1.1$ meV. The cavity-exciton detuning is set to $\Delta = -1$ meV.

These numerical results can be explained by applying the Weiskopf-Wigner approximation to the *X-X* correlation term entering the equation for the probe polarization P_1. Assuming the two pulses to be sufficiently spectral narrow and centerd to excite only one polariton mode ($\omega_j = \omega_U, \omega_L$) we obtain,

$$(\Delta_I - i\Gamma_I)P_1, \quad (1)$$

with

$$\Delta_I = |P_2|^2 \left[\beta + \wp \int \frac{F(\omega)}{\omega_1 + \omega_2 - \omega} d\omega \right] \quad (2)$$

and

$$\Gamma_I = |P_2|^2 \pi F(\omega_1 + \omega_2), \quad (3)$$

where $F(\omega)$ is the spectral density of the *X-X* correlation. As expected Eqs. (1–3) show that the X-X correlation in SMCs determines an intensity-dependent absorption mechanism and a renormalization of the mean-field interaction b determined by the spectrum of four-particle correlations. In particular Γ_I is directly proportional to the spectral density of the *X-X* correlation function calculated at $\omega_1 + \omega_2$.

We observe that by tuning the exciton-cavity detuning, it is possible to obtain information of $F(\omega)$ over a spectral region larger than 40 meV contered on twice the bare-exciton energy. As expected and as confirmed by exact numerical calculations[6] $F(\omega)$ displays strong variations within this spectral region. These variations fully explain the results in Fig. 1 and the corresponding experimental findings.[1]

This should open the way to the spectroscopy of four-particle correlations in SMCs.

References

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