Transient Optical Stark Shift and Excitation Induced Dephasing in the Non-Perturbative Regime

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We show that the exciton–exciton scattering continuum has a significant influence on the transient non-linear optical response of semiconductor microcavities. Comparison between transient shifts observed in pump and probe experiments and calculations demonstrates almost complete cancellation between mean-field and two-exciton continuum contributions. Moreover, we show that suppression of non-linear absorption of the lower polariton observed in many experiments is determined by the specific spectral shape of these correlations. In particular, we find that the pump induced dephasing of polariton resonances is proportional to the spectrum of four-particle correlations calculated at the energy sum of the pumped and probed polaritons.

Studies over the past decade have shown that Coulomb correlations between excitons dominate the non-linear optical response of semiconductors, in marked contrast to the behaviour of atomic systems [1]. These Coulomb correlations and other many-body interactions are important not only for semiconductors, but also for all condensed-matter systems.

In this paper we show that the scattering continuum after the exciton–exciton correlation has a significant influence on the transient non-linear optical response of semiconductor microcavities, and we demonstrate that semiconductor microcavities form an ideal laboratory for investigating Coulomb correlations. In particular, we show that routinely used optical techniques, such as transient pump and probe spectroscopy, can provide a direct measurement of the low energy tail of the spectrum of four-particle correlations.

We start from the equations for the third-order exciton polarization and cavity field describing quantum optical effects and coherent non-linear optics in semiconductor microcavities [2], and perform the semiclassical factorization. Then we express the non-linear term coming from Coulomb interaction as an exciton–exciton (X–X) mean-field interaction plus a correlation term which is expressed as a two-exciton correlation function [3] and apply this procedure to the non-perturbative regime [2]. Finally we include multiple scattering simply by replacing in the non-linear sources the linear polarization and light fields with the total fields. Multiple scattering processes are expected to be very effective in cavity embedded QWs due to multiple reflections at the Bragg mirrors.

In order to keep the treatment as simple as possible, we consider a specific configuration with a probe beam 1 sent along the growth axis of the microcavity (in-plane wave

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vector $\mathbf{k} = 0$) and a slightly tilted ($\mathbf{k} \simeq 0$) pump beam 2 with the same circular polarization of the probe. The coherent dynamics of the photon–exciton system can be described by a set of coupled equations of motion for the probe and for the pump polarization densities P_j and intracavity fields E_j ($j = 1, 2$). In particular the time evolution of the probe signal is determined by the equations

$$\frac{\partial}{\partial t} E_1 = -(\gamma_c + i\omega_1) E_1 + iVP_1 + \sqrt{g_c} E_1^{in}, \tag{1}$$

$$\frac{\partial}{\partial t} P_1 = -(\gamma_x + i\omega_x) P_1 + iVE_1 - i\Omega_1^{(NL)}, \tag{2}$$

where ω_j and ω_x, γ_c and γ_x are the energies and dephasing rates of cavity photons and QW excitons, V is the collective dipole coupling rate, E_j^{in} describes the input light field, and g_c gives the fraction of input photons passing the cavity mirror. The relevant nonlinear source term is given by $\Omega_1^{(NL)} = \Omega_1^{(a)} + \Omega_1^{(b)}$, where the first term originates from phase-space filling,

$$\Omega_1^{(a)} = +\frac{1}{n_c} (|P_2|^2 E_1(t) + P_2^* P_1 E_2), \tag{3}$$

where n_c is the exciton saturation density, and $\Omega_1^{(b)}$ comes from the Coulomb interaction between electrons and can be written in the form

$$\Omega_1^{(b)} = +\beta |P_2|^2 P_1 - iP_2^* \int_{-\infty}^{t} F(t-t') P_1(t') P_2(t') \, dt', \tag{4}$$

where β is a measure of the mean-field X–X interaction and $F(t)$ is the retarded memory function containing the equal-spin exciton–exciton correlation [3, 4]. An analogous set of equations can be derived for the pump dynamics simply by the replacement $1 \rightarrow 2$ in Eqs. (1) and (2), and with the relevant non-linear sources given by

$$\Omega_2^{(a)} = \frac{1}{n_c} |P_2|^2 E_2(t), \tag{5}$$

$$\Omega_2^{(b)} = \frac{1}{2} \left(\beta |P_2|^2 P_2 + P_2^* \int_{-\infty}^{t} F(t-t') P_2(t') P_2(t') \, dt' \right). \tag{6}$$

These equations can be readily generalized to include other polarization configurations and excitation geometries.

Before presenting numerical calculations based on the above equations, it is useful to perform an analytical study of the two-exciton correlation in the non-perturbative regime. Let us consider a pump and a probe ($j = 1, 2$) ultrafast pulse with zero time delay, each centered on the lower or upper polariton energy. For the sake of simplicity, the two pulses are assumed to be sufficiently spectrally narrow to excite only one (the lower at ω_L or the upper at ω_U) normal mode. The excited pump and probe polarizations can be written as $P_j = \Theta(t) X_j(t) e^{-i\omega_j t}$, where ω_j is the energy of the polariton mode excited by the j-th beam and $X_j(t)$ are polarization amplitudes, which are slowly varying functions of time as compared to $e^{-i\omega_j t}$ and to the decay time of the memory function $F(t)$. This assumption is analogous to the Weisskopf-

Wigner approximation adopted to analyze spontaneous emission in two-level transitions. We obtain

$$\Omega_1^b = (\Delta_I - i\Gamma_I)\,|P_2|^2\,P_1\,, \tag{7}$$

with

$$\Gamma_I = \pi\mathcal{F}(\omega_1+\omega_2)\,, \tag{8}$$

$$\Delta_I = \beta + \wp\int \frac{\mathcal{F}(\omega)}{\omega_1+\omega_2-\omega}\,\mathrm{d}\omega\,. \tag{9}$$

Equations (7)–(9) show that the X–X correlation in SMCs determines an intensity dependent dephasing mechanism and a renormalization of the mean-field interaction β. As expected they are proportional to the coherent exciton density generated by the pump beam $|P_2|^2$. They also have an energy dependence determined by the spectrum of four-particle correlations. We observe that by tuning the relative energy Δ between cavity and exciton resonances, and in addition exploiting the three possibilities $\omega_1+\omega_2 = 2\omega_U$, $\omega_U+\omega_L$, $2\omega_L$, it is possible to span Γ_I and hence $\mathcal{F}(\omega)$ over a spectral region centered on $2\omega_0$ larger than 40 meV for typical III–V SMCs. As expected and as confirmed by exact numerical calculations for a one-dimensional semiconductor model [4], $\mathcal{F}(\omega)$ displays strong variations within this spectral region. In order to describe the effects of two-exciton correlations, we adopt a model correlation function introduced by Östreich and Sham [5] which combines a low-frequency power law with a high-frequency decay approximated as exponential. In Fig. 1a the spectral density of the equal-spin correlation function Γ_I is plotted by imposing a quadratic low-frequency behaviour and by using $\beta = 49/\pi a^2$ meV and $F(0) = \beta^2/1.5$ (a is the two-dimensional X Bohr radius). The zero of energy corresponds to the energy of two non-interacting 1s excitons ($2\omega_0 = 0$). Figure 1a together with Eq. (8) clearly predicts a suppression, at zero detuning, of the power dependent absorption for $\omega_1+\omega_2 < 0$, in other words when $\omega_1+\omega_2 = 2\omega_L$. This suppression has been observed by different groups [6–10]. As shown in Fig. 1b, also the effective non-linear susceptance Δ_I displays a relevant frequency dependence passing from positive values (blue-shift) at negative energies to

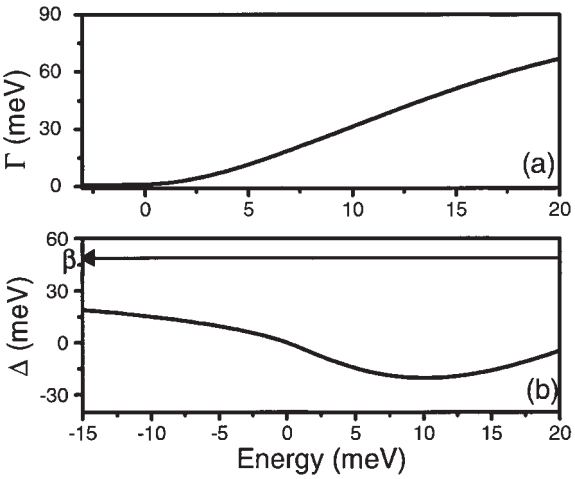

Fig. 1. a) Spectral density of the equal-spin correlation function, parameters are given in the text. b) Non-linear shift as a function of energy

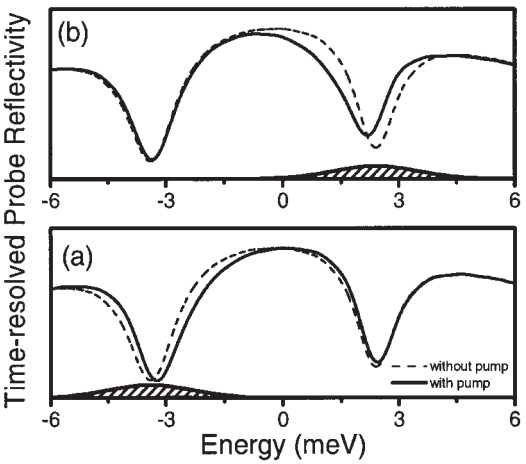

Fig. 2. Spectra of the calculated time-resolved probe reflectivity with the pump switched on (solid line) and off (dashed line) for narrow-band pump pulses centered on the a) lower and b) upper polariton energy. We have used the dipole coupling rate $V = 2.85$ meV, exciton dephasing rate $\gamma_0 = 0.35$ meV, and photon escape rate $\gamma_c = 1.1$ meV, and we have adopted the model correlation function described in the text

negative values (red-shift) at positive energies. Furthermore, Δ_I is almost zero at $\omega = 0$ owing to cancellation effects imposed by a sum rule and included in the model [4, 5]. We have to point out that, while the suppression of non-linear absorption and these almost complete cancellation effects are independent of the specific model of correlations, the calculated shifts particularly at positive energy can significantly depend on the specific model adopted. Very recently a microscopic calculation of the X–X correlation in two dimensions has been presented [11]. The obtained low energy spectrum is different from the model adopted here, and determines energy shifts that can be rather different from those obtained with the model adopted here. Thus, results shown in Fig. 1b have to be taken just as indications of how transient shifts induced by a pump beam can provide detailed information on the two-exciton correlations. Figure 1b predicts a power dependent shift of the polariton peaks passing from blue to red according to the sign of $\omega_1 + \omega_2$. Thus, shifts and non-linear absorption measured in simple pump and probe configurations in SMCs are expected to provide precise information on the spectrum of two-exciton correlations and to test different theoretical models and approximations.

In Fig. 2 we display spectra of the calculated time-resolved probe reflectivity with and without the presence of the pump beam. We use a Gaussian 700 fs pump pulse with zero time delay with respect to the probe pulse producing a maximum exciton density $n \simeq 0.1/\pi a^2$. In order to discriminate the energy dependence of power dependent absorption and shifts, the spectrum of the pump beam is centered on the lower or upper polariton energy. The probe is a 100 fs pulse 10^{-4} weaker than the 100 fs pump pulse centered on ω_0. When the lower polariton is pumped (Fig. 2a), it experiences a blueshift without a corresponding shift of the upper line in agreement with the Weisskopf-Wigner approximation. Furthermore, no reduction (due to non-linear absorption) of the lower line is observed, while the upper resonance suffers a small bleaching. When the upper line is pumped (Fig. 2b), it redshifts leaving the lower line almost unmoved. In addition, a significant non-linear absorption of the upper line and a very small non-linear absorption of the lower line can be observed. These puzzling behaviours of absorption and shifts, determined by the X–X correlation, are fully consistent with the shifts and absorption rates obtained within the Wigner-Weisskopf approximation (see Figs. 1a and b).

In conclusion we have investigated the role of four-particle correlations in the ultrafast time-resolved non-linear response of SMCs. We have shown that they can affect significantly the transient non-linear optical properties of SMCs. We have clarified the origin of the observed very different non-linear absorption rates of the two polariton branches showing that they reflect the spectral properties of the two-exciton scattering continuum. Furthermore, the analytical study and the numerical results presented here demonstrate that the non-perturbative regime of SMCs provides a unique tool for accessing the spectrum of four-particle correlations.

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