Quantum Theory of Nonlinear Semiconductor Microcavity Luminescence Explaining "Boser" Experiments

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Photoluminescence experiments from semiconductor quantum wells inside a microcavity are reported which exhibit a thresholdlike transition already below lasing. A fully quantum mechanical theory for an interacting system of photons and Coulomb-correlated quantum well electrons and holes inside a microcavity is presented. The experimental results previously attributed to bosonic condensation are explained consistently in terms of fermionic electron-hole correlations. [S0031-9007(97)04762-5]

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Light matter interaction effects in semiconductor microcavities have been actively studied since the manifestation [1] of excitonic normal mode coupling (NMC) in the nonperturbative regime. Such interactions in atomic [2] and molecular [3] cavity systems are rather well understood on the basis of models related to discrete level systems. A similar approach for semiconductors describes the optical properties with excitonic bound states using a bosonic approximation [4,5]. Thus the full electron-hole Coulomb interaction in the many-body Hamiltonian is approximated by including only some aspects of the interband attractive part.

Recent experiments show that the photoluminescence (PL) properties of semiconductor microcavities contain an unexpected transition [6]: When the cavity mode is energetically above the exciton, the intensity of the higher energy PL peak exhibits a thresholdlike behavior, eventually overtaking the lower energy PL peak. This pronounced effect has been widely proclaimed as an example of "boser action" [6] being related to Bose condensation [5]. Our own experiments reproduce this striking behavior. However, we go further and show that NMC is collapsing as the PL crosses over, eliminating the condensation hypothesis.

To clarify the "boser problem" from a theoretical point of view, one needs a quantum theory for the microcavity luminescence that does not make a bosonic approximation for the Coulomb-correlated electron-hole pairs. Clearly, such a fully quantum mechanical analysis of an interacting photon-semiconductor electron-hole system poses a considerable challenge to current theories. A well-established approximation scheme exists in the semiclassical regime, where the major difficulties arise from the consistent inclusion of the carrier-carrier Coulomb interaction effects [7,8]. The simplest self-consistent approximation scheme leads to the Hartree-Fock semiconductor Bloch equations for which systematic improvements have been published and analyzed [9–13]. These theories have been used successfully, e.g., to explain the density dependent saturation of the microcavity normal mode coupling resonances [14], as well as the ultrafast pulse propagation dynamics and the associated excitation induced dephasing [15].

In this Letter, we present a major extension of previous theories by developing a fully quantum mechanical analysis of the interacting electron-hole photon system that includes both attractive and repulsive Coulomb effects. This theory, evaluated for studying the microcavity PL, reproduces consistently the following observations: In the transitions from NMC to weak coupling, the PL remains double peaked, then becomes single peaked, and then lases. Since electrons and holes are treated correctly as fermions, we can check for the validity of a bosonic approximation finding that it is already clearly inappropriate for excitation levels below the "boser transition."

The field is quantized using an expansion into empty cavity modes $u_{\mathbf{q}}$ associated with a photon destruction operator $b_{\mathbf{q}}$, where $\mathbf{q} = (\mathbf{q}_{||}, q)$ is the wave vector. Since the luminescence detection outside the cavity is performed in the normal direction, it is sufficient to analyze only the modes $\mathbf{q}_{||} = \mathbf{0}$. The coupling of the light to the semiconductor electron-hole pairs is described by the dipole-interaction Hamiltonian [16]. The Coulomb interaction between electrons and holes is included by the standard two-band many-body Hamiltonian [8].

We denote the fermion operators for conduction and valence band electrons with momentum $\hbar\mathbf{k}$ by $c_{\mathbf{k}}$ and $v_{\mathbf{k}}$, respectively. To obtain the quantum form of the semiconductor Bloch equations, we follow the standard procedure [8] and evaluate the Heisenberg equations for the microscopic polarization $\hat{P}_{\mathbf{k}} = v_{\mathbf{k}}^{\dagger} c_{\mathbf{k}}$, the carrier occupations $\hat{n}_{\mathbf{k}}^{c} = c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}}$, $\hat{n}_{\mathbf{k}}^{v} = v_{\mathbf{k}}^{\dagger} v_{\mathbf{k}}$, and, additionally, also for the photon operator b_q. The resulting operator equations can be written as

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$$i\hbar \frac{\partial}{\partial t} \hat{P}_{\mathbf{k}} = (\epsilon^c_{\mathbf{k}} - \epsilon^v_{\mathbf{k}})\hat{P}_{\mathbf{k}} + \mu_{cv}(\mathbf{k})(c^\dagger_{\mathbf{k}}\tilde{E}c_{\mathbf{k}} - v^\dagger_{\mathbf{k}}\tilde{E}v_{\mathbf{k}}) + \sum_{\mathbf{k}'\mathbf{k}''} V(\mathbf{k}' - \mathbf{k}) \times [v^\dagger_{\mathbf{k}}(c^\dagger_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}}c_{\mathbf{k}''} + v^\dagger_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}}v_{\mathbf{k}''})c_{\mathbf{k}'} - v^\dagger_{\mathbf{k}'}(c^\dagger_{\mathbf{k}''}c_{\mathbf{k}'+\mathbf{k}''} + v^\dagger_{\mathbf{k}''}v_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}})c_{\mathbf{k}}], \quad (1)$$

$$i\hbar \frac{\partial}{\partial t} \hat{n}^c_{\mathbf{k}} = \left[\mu_{cv}(\mathbf{k})v^\dagger_{\mathbf{k}}\tilde{E}c_{\mathbf{k}} - \sum_{\mathbf{k}',\mathbf{k}''} V(\mathbf{k}' - \mathbf{k}) \times c^\dagger_{\mathbf{k}'}(c^\dagger_{\mathbf{k}''}c_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}} + v^\dagger_{\mathbf{k}''}v_{\mathbf{k}'+\mathbf{k}''-\mathbf{k}})c_{\mathbf{k}} - \text{H.c.} \right], \quad (2)$$

$$i\hbar \frac{\partial}{\partial t} b_q = \hbar\omega_q b_q + i\mathcal{E}_q \tilde{u}^*_q(\hat{P} + \hat{P}^\dagger), \quad (3)$$

where $\hat{P} = \sum_{\mathbf{k}} \mu^\star_{cv}(\mathbf{k})\hat{P}_{\mathbf{k}}$ and μ_{cv} is the dipole matrix element. Furthermore, $\frac{\partial}{\partial t}\hat{n}^v_{\mathbf{k}} = -\frac{\partial}{\partial t}\hat{n}^c_{\mathbf{k}}$, $V(\mathbf{k})$ is the quantum well (QW) Coulomb matrix element, $\mathcal{E}_q$ is the vacuum field amplitude, $\omega_q = cq$, and $\epsilon^{c,v}_{\mathbf{k}}$ are the free-particle energies, respectively. In these equations, $\tilde{E} = \int g(z)E(z)\,dz$ is the effective QW field, $\tilde{u}_q = \int g(z)\,u_q(z)\,dz$, $u_q(z)$ is the cavity mode function, and $g(z)$ is the QW confinement factor incorporating the single-particle envelope functions. In Eqs. (1)–(3), all operators are always normally ordered. In the semiclassical limit, $\langle c^\dagger_{\mathbf{k}}Ec_{\mathbf{k}}\rangle = \langle c^\dagger_{\mathbf{k}}c_{\mathbf{k}}\rangle\langle E\rangle$, our equations reduce to the semiconductor Bloch equations [7,8,17].

The operator, Eqs. (1)–(3), serves as a general starting point to investigate quantum correlations in semiconductor microcavity systems. In this Letter, we apply Eqs. (1)–(3) to analyze the microcavity PL experiments for various electron-hole pair densities. In the absence of a classical driving field, the light field and carrier polarization are mutually incoherent, i.e., $\langle b_q\rangle = 0$ and $\langle\hat{P}_{\mathbf{k}}\rangle = 0$. Hence, the lowest order nonzero quantities are $f^e_{\mathbf{k}} = \langle\hat{n}^c_{\mathbf{k}}\rangle$, $f^h_{\mathbf{k}} = 1 - \langle\hat{n}^v_{\mathbf{k}}\rangle$, and the correlations $\langle b^\dagger_q\hat{P}_{\mathbf{k}}\rangle$, and $\langle b^\dagger_q b_{q'}\rangle$. To truncate the hierarchy of the resulting equations of motion, we use the dynamical decoupling scheme [7,8] for the four carrier operator terms and the mixed photon-carrier operator terms. This way we obtain a closed set of semiconductor luminescence equations,

$$i\hbar \frac{\partial}{\partial t} \langle b^\dagger_q\hat{P}_{\mathbf{k}}\rangle = (-\tilde{\epsilon}^c_{\mathbf{k}} - \tilde{\epsilon}^v_{\mathbf{k}} - \hbar\omega_q + E_G)\langle b^\dagger_q\hat{P}_{\mathbf{k}}\rangle + (f^e_{\mathbf{k}} + f^h_{\mathbf{k}} - 1)\Omega(\mathbf{k},q) + f^e_{\mathbf{k}}f^h_{\mathbf{k}}\Omega_{SE}(\mathbf{k},q), \quad (4)$$

$$i\hbar \frac{\partial}{\partial t} \langle b^\dagger_q b_{q'}\rangle = \hbar(\omega_{q'} - \omega_q)\langle b^\dagger_q b_{q'}\rangle + i\mathcal{E}_q\tilde{u}_q\langle b_{q'}\hat{P}_H\rangle + i\mathcal{E}_{q'}\tilde{u}^*_{q'}\langle b^\dagger_q\hat{P}_H\rangle, \quad (5)$$

$$i\hbar \frac{\partial}{\partial t} f^{e(h)}_{\mathbf{k}} = 2i\,\mathrm{Im}[\mu^*_{cv}(\mathbf{k})\langle\hat{P}_{\mathbf{k}}\tilde{D}\rangle], \quad (6)$$

where $\tilde{\epsilon}^{c,v}_{\mathbf{k}}$ are the renormalized single-particle energies, $\tilde{D} = \int g(z)D(z)\,dz = \sum_q i\mathcal{E}_q\tilde{u}_q b_q + \text{H.c.}$, $\hat{P}_H = \hat{P} + \hat{P}^\dagger$, and $\tilde{E} - \tilde{D} \propto \hat{P}_H$. In Eq. (4), the term containing the renormalized field $\Omega(\mathbf{k},q) = \mu_{cv}(\mathbf{k})\langle b^\dagger_q\tilde{E}\rangle + \sum_{\mathbf{k}'} V(\mathbf{k}' - \mathbf{k})\langle b^\dagger_q\hat{P}_{\mathbf{k}'}\rangle$ describes the stimulated emission or absorption and the term including $\Omega_{SE}(\mathbf{k},q) = \mu_{cv}(\mathbf{k})i\mathcal{E}_q\tilde{u}_q$ leads to spontaneous emission, respectively. Both, the spontaneous and stimulated terms act as sources for the field-particle correlations $\langle b^\dagger_q\hat{P}_{\mathbf{k}}\rangle$ which enter in Eqs. (5) and (6). Note that, without the stimulated term containing the Coulomb renormalized field, NMC could not be obtained. The equations for modes $\mathbf{q}_{||} \neq \mathbf{0}$ are analogous to Eqs. (4) and (5). However, as long as carrier density depletion can be neglected, modes with different $\mathbf{q}_{||}$ do not couple since in-plane momentum is conserved.

Before we present a numerical evaluation of the theory, we briefly discuss the main observations from our experimental studies. We use a QW microcavity consisting of a $3/2\lambda$ GaAs spacer between symmetric 99.6% GaAs/AlAs Bragg mirrors, with two 8 nm $In_{0.04}Ga_{0.96}As$ QWs placed at the antinodes of the intracavity field. The cavity mode is tuned with respect to the exciton resonance by scanning across the tapered sample. The sample was held at a temperature of 4 K and excited in a reflection minimum at 780 nm by 0.5 μs pulses (practically cw) with a 10% duty cycle to reduce heating. The PL is collected in the normal direction from a small solid angle, $4\pi \times 10^{-3}$ sr, and imaged onto an aperture which has an area an order of magnitude smaller than the image of the PL spot. Thus, only PL coming from the central, most uniformly excited region is collected.

The measured PL spectra are plotted in the left column of Fig. 1. We see that with increasing excitation power the upper energy peak overtakes the lower one as shown also in Fig. 2(a); this is the boser action reported previously [6]. Because the PL is still double peaked after the crossing, it has been argued that NMC still exists (giving possibility to Bose condensation). However, Fig. 2(c) shows that above crossover the splitting no longer decreases with increasing intensity; the splitting has already reduced from the generalized Rabi frequency value to simply the detuning. Lasing eventually occurs very close to the position of the upper peak at crossover. Moreover, transmission spectra obtained for the same pump conditions as the PL spectra show that just above crossover the transmission becomes single peaked. Also, the crossover power is more than half the lasing threshold power (Fig. 3), which already indicates a rather high density for boser action. Furthermore, the boser

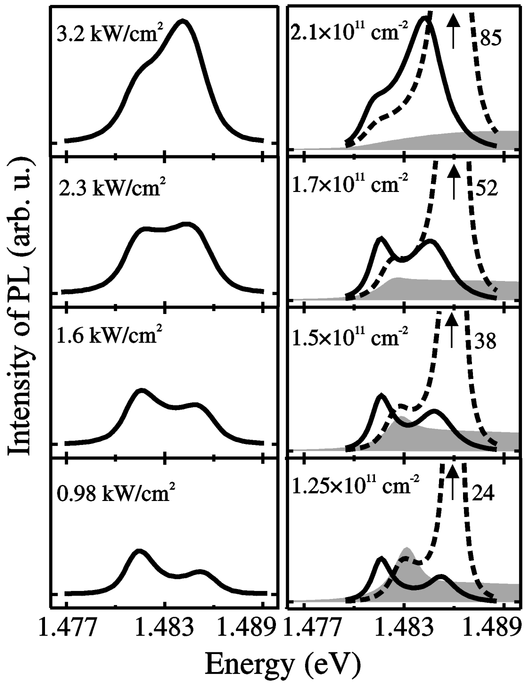

FIG. 1. In the left column, the measured nonlinear cw PL spectra are plotted for various excitation intensities and detuning +3 meV. The right column shows the computed results of the full theory (solid line) and a computation excluding NMC (dashed line) for various excitation densities; the same arbitrary units are used for all densities. The maximum value of the high energy peak is indicated by the number at the arrow. The shaded curves are the computed excitonic absorption spectra.

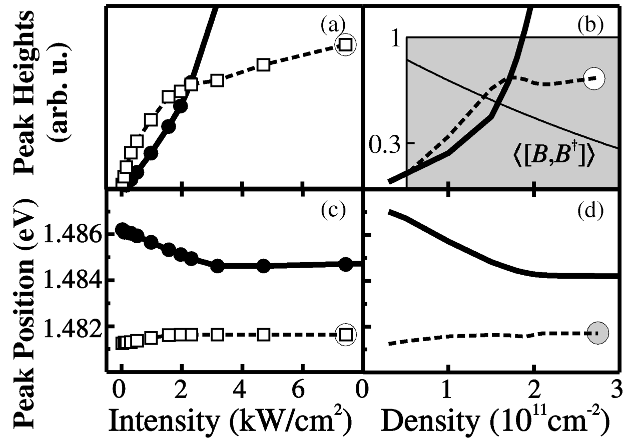

FIG. 2. Measured microcavity PL peak intensities (a) and peak energies (c) vs cw excitation intensity for the high energy peak (solid line) and the low energy peak (dashed line) corresponding to Fig. 1. Results of the microscopic theory are shown in (b) and (d). The circle denotes the highest excitation where the low energy peak can be resolved. Inset to (b) shows the calculated expectation value of the Bose commutator vs carrier density.

action region cannot show coherent emission since its efficiency for perpendicular emission is much lower than for lasing. For the pulsed excitation, the time-integrated PL includes significant averaging in comparison to cw spectra at constant density. Thus the double peaked region seems to last longer than in the cw case because the lower peak PL is enhanced as the carrier density decays away. [18]

To analyze the experiments, Eqs. (4)–(6) are solved numerically starting from zero correlations. Assuming that $f_{\mathbf{k}}^{e(h)}$ is a constant quasiequilibrium Fermi-Dirac distribution, we only have to evolve Eqs. (4) and (5) to the steady state. We compute the PL spectrum which is proportional to the flux passing the detector $\frac{d}{dt}\langle b_q^\dagger b_q \rangle$. Carrier scattering is included via the dephasing rate γ whose density dependent value is computed with an independent quantum kinetic calculation [13,14]. For standard material parameters corresponding to the experiment, we obtain the results in the right column of Figs. 1 and 2. Clearly, our fully quantum mechanical theory is capable of reproducing the experimental observations. To demonstrate that our computed results do not rely on the bosonic nature of the excitation and condensation effects, we have plotted in Fig. 2(b) the expectation value of the commutator between two exciton operators [8], $B = \sum_{\mathbf{k}} \psi_0(\mathbf{k}) v_{\mathbf{k}}^\dagger c_{\mathbf{k}}$ and $B^\dagger$, where $\psi_0(\mathbf{k})$ is the Fourier transform of the $1s$-exciton wave function. This commutator equals unity in the Bose limit and has to be very close to unity in order to reasonably approximate excitons as Bosons. However, our results inevitably show that, for the elevated carrier densities at which the PL peaks cross, the commutator varies between 0.7 and 0.3, i.e., far below the ideal bosonic value of 1. Hence, at crossover of the PL peaks, the density is so high that the transition from nonperturbative normal mode coupling to perturbative weak coupling is almost complete, clearly excluding the possibility of Bose condensation effects and the application of simple bosonic models.

To explain the true microscopic origin of the boser action, we compute the PL spectrum where we artificially

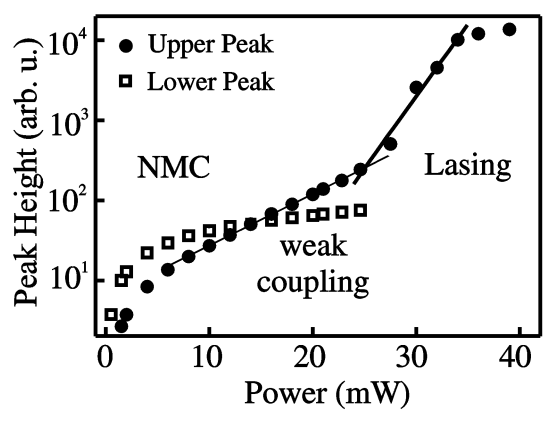

FIG. 3. Peak PL intensity vs pump power for +4.2 meV detuning and 100 fs pulses exciting above the cavity stop band. The transition from normal mode coupling (NMC) to weak coupling occurs in the boser action regime.

switch off the unrenormalized stimulated field term $\mu_{cv}(\mathbf{k})\langle b_q^\dagger \tilde{E}\rangle$, thus effectively decoupling PL and NMC. The results for various carrier densities are plotted in Fig. 1 as dashed lines. We obtain double peaked emission, where one peak is always at the excitonic resonance and one is at the empty cavity mode. Without NMC, the effect of spontaneous emission is basically that of a classical emitter inside a detuned cavity. Increasing the emission intensity leads to a dramatic increase of the emission at the cavity mode, but only to a weak increase of the emission at the frequency of the detuned emitter. The frequency of the detuned emitter remains close to the zero density exitonic resonance even for densities where the excitonic *absorption* (shaded curves in Fig. 1) peak has been completely bleached.

The coupled set of Eqs. (4)–(6), including the stimulated field term, is necessary to explain the experimental results. Since the high energy peak of the PL is energetically close to the unrenormalized semiconductor band gap, it is strongly suppressed for low excitation densities and therefore the low energy peak dominates. For higher carrier densities, the semiconductor absorption is gradually bleached (Fig. 1), reducing the suppression of the high energy luminescence, eventually allowing the high energy peak to grow considerably. Since both effects, increased spontaneous emission and decreased semiconductor absorption, strengthen the high energy peak, the heights of the two PL peaks cross. Consequently, boser action is explained as nonlinear semiconductor dynamics of an interacting electron-hole system without “bosonic effects.” After the peak crossing, NMC is gone since the excitonic absorption resonance is bleached. However, the PL is still double peaked because of the detuned emitter situation discussed above [19].

In summary, a many-body treatment for the coupled system of quantum well carriers interacting with the quantized light field in a semiconductor microcavity has been presented. The experimentally observed nonlinear PL characteristics are reproduced and explained within the framework of transitions from normal mode coupling to weak coupling double peaked emission (instead of boser [6]) and lasing. The full quantum theory outlined here allows us to study not only luminescence and lasing properties in detail but also to investigate higher order photon correlations in semiconductor microcavity systems.

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