Crossover from Exciton to Biexciton Polaritons in Semiconductor Microcavities

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Pump-probe measurements in a microcavity containing a quantum well show that a population of circularly polarized (σ^+) excitons can completely inhibit the transition to σ^- one-exciton states by transferring the oscillator strength to the biexcitonic resonance. With increasing pump intensity the linear exciton-polariton doublet evolves into a triplet polariton structure and finally into a shifted biexciton-polariton doublet. A theoretical model of interacting excitons demonstrates that the crossover from exciton to biexciton polaritons is driven by three-exciton Coulomb correlation.

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Coulomb correlation in a gas of interacting electron-hole pairs is one of the most intriguing and fundamental topics in the domain of condensed matter physics. Concerning optical properties of semiconductors, the exciton resonance is the dominant electron-hole correlation effect when the light interacting with the sample is nearly resonant with the band gap. Considering the correlation to higher orders, the exciton-exciton interactions can give rise to the formation of the excitonic molecule, namely, the biexciton [1–3]. A great effort has been made to understand how the polarization of two-exciton states influences the nonlinear polarization in experiments like four-wave mixing, hyper-Raman scattering, two-photon absorption, mostly focusing on the coherent regime [4–8]. In the opposite limit of the incoherent regime, when the polarization induced by the laser excitation is lost, the exciton to biexciton transition induced by an optical probe is driven only by the exciton population. In this physical situation, a major issue is the biexcitonic correlation induced by a dense σ^+ exciton population on the σ^- transition. Up to now experiments on bare quantum wells (QWs) [9] and on microcavities [10] have shown a continuous redshift of the σ^- exciton energy with growing σ^+ population. The high scattering rate induced by the dense σ^+ population prevented the resolution of a biexcitonic peak in the σ^- spectrum, as the exciton line broadening was exceeding the biexciton binding energy ($\simeq$2 meV). The corresponding models treated the exciton-exciton interaction in a mean field approximation, thus reproducing the continuous shift (see, e.g., [11]).

In this Letter, we present measurements on a microcavity containing a QW. The exciton to biexciton transition is resolved in the incoherent regime, thus enabling a detailed study of the exciton and biexciton oscillator strengths for growing excitation densities. We report an oscillator strength transfer from the excitonic to the biexcitonic transition, i.e., the excitonic absorption can be quenched by the rise of the biexcitonic absorption. The sample is excited by intense circularly polarized (σ^+) pump pulses and probed at different delays by a weak countercircularly polarized (σ^-) test beam. In this way the optical nonlinearities observed in the σ^- transition are due entirely to Coulomb correlation with the σ^+ population and the effects of phase space filling are ruled out. Our experimental results show that the presence of σ^+ excitons in sufficient density can inhibit the transition to the σ^- exciton states by *transferring* the oscillator strength to the biexciton transition. This transition is strongly coupled to the cavity photon mode. In analogy with the exciton polaritons [12], we call the mixed states biexciton polaritons. By varying the optical excitation density, the crossover from exciton to biexciton polaritons can be monitored. In our high finesse microcavity the cavity photon mode is spectrally very narrow ($\simeq$0.1 meV FWHM) and it is chosen to be resonant with the lowest exciton state. The excitation of free electron-hole pairs is highly suppressed and the exciton-free carrier scattering, which is the main source of broadening in the high excitation regime, is considerably reduced. As the exciton molecule formation appears to be the dominant feature in the experiments, the theoretical analysis has to go beyond a mean field treatment of the exciton-exciton interactions. The data are compared with a model of interacting excitons where the Coulomb correlation is included up to the third order. The σ^- probe susceptibility is analytically calculated in the presence of an incoherent σ^+ exciton population. The obtained QW susceptibility satisfies a *sum rule:* the total oscillator strength of the exciton and biexciton transitions does not depend on the density of σ^+ excitons. The solution of the Maxwell equations for the microcavity with the calculated QW susceptibility leads to transmission spectra that are in excellent agreement with the experiments.

The sample, grown by molecular-beam epitaxy, is a single 7.5-nm-wide $In_{0.03}Ga_{0.97}As$ QW embedded in a GaAs wedge shaped λ cavity, whose mirrors are $AlAs/AlGa_{0.9}As_{0.1}$ distributed Bragg reflectors. The linear characterization reveals a Rabi splitting of 3.6 meV at resonance (Fig. 1a); the average linewidth of the two polaritons is 0.13 meV [13].

We set up a degenerate pump and probe experiment with nearly transform limited $\simeq$100-fs-long pulses. The sample was kept at a temperature of 2 K in a superfluid helium bath. The evolution of the probe transmission spectrum with increasing pump intensity is shown in Fig. 1b. The probe pulse, countercircularly polarized with respect to the pump, tests the sample 6 ps after excitation, when the population created by the pump is still present, but has already lost its coherence.

In the linear regime, the probe transmission spectrum exhibits the Rabi splitting of the exciton polaritons (the polariton linewidth is determined by the spectral resolution of the detection apparatus). For increasing excitation rate, the Rabi doublet transforms into a triplet. The appearance of three resonances is an unambiguous proof that two electronic transitions are coupled to the cavity photon mode. The three peaks evolve differently as the pump density increases. The lowest resonance, rising approximately at the energy of the biexciton transition (just below the unperturbed lower exciton polariton), has essentially the character of a biexciton polariton. It redshifts for increasing excitation intensities. Also the uppermost polariton redshifts and becomes dominant at the highest pumping rates. Conversely the central peak of the triplet loses intensity and blueshifts towards the bare exciton energy; it therefore has mainly the character of an exciton polariton. Further, redshift and splitting of the biexciton polaritons saturate when the central line disappears. From this behavior we deduce that with increasing σ^+ exciton density the oscillator strength of the biexciton transition increases at the expense of that of σ^- exciton.

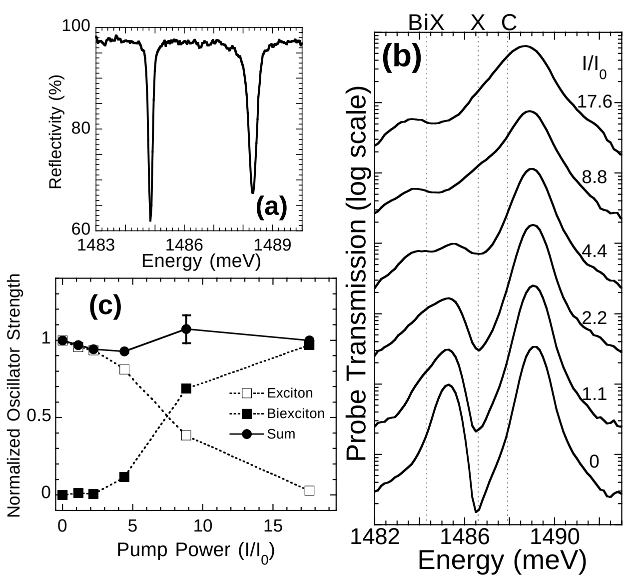

FIG. 1. (a) Continuous wave reflection spectrum at cavity to exciton resonance, measured at a temperature of 4 K, spectral resolution $\simeq$0.1 meV (courtesy of R. P. Stanley). (b) Transmission spectra for growing pump intensities I at a 6 ps delay (spectral resolution $\simeq$ 0.7 meV); the three dashed lines mark the energies of the empty-cavity mode (C), the bare exciton (X), and biexciton (BIX) transitions; $I_0 \equiv 10^{12}$ photons $\mathrm{cm}^{-2}\,\mathrm{pulse}^{-1}$. (c) Normalized oscillator strengths of the exciton and biexciton transitions and their sum as a function of the pump intensity. A 0.1-meV incertitude in reading the polariton energies gives an error in the determination of the oscillator strength of about 10%.

The exciton transition (energy E_X), biexciton transition (E_{BIX}), and cavity mode (E_C) are a system of three coupled oscillators. The three eigenenergies $P_{1,2,3}$ are the solutions of the secular equation: $(E - E_C)(E - E_X)(E - E_{BIX}) - |V|^2 \tilde{f}_X (E - E_{BIX}) - |V|^2 \tilde{f}_{BIX} (E - E_X) = (P_1 - E) \times (P_2 - E)(P_3 - E) = 0$, where V is the half Rabi splitting in the linear regime and $\tilde{f}_{X,BIX}$ are the exciton and biexciton oscillator strengths normalized to the excitonic oscillator strength in the linear regime. In fact E_C and E_X are known from the sample characterization, E_{BIX} measured in the bare quantum well is 2 meV, and $P_{1,2,3}$ are measured in the transmission spectra (Fig. 1b). Therefore $\tilde{f}_{X,BIX}$ can be calculated by inverting the secular equation. In Fig. 1c $\tilde{f}_{X,BIX}$ are plotted as a function of the pump intensity: the exciton oscillator strength decreases and conversely the biexcitonic oscillator strength increases. Remarkably the total oscillator strength of the two transitions is constant during the crossover: the biexciton transition takes the oscillator strength from the excitonic one.

The spectra taken varying the pump to probe delay support and confirm our interpretation. Two different regimes can be distinguished in the temporal dynamics of intense excitations in microcavities. In the coherent regime the pump drives deep Rabi oscillations of the exciton density [14,15]; once coherence is lost, a monotonic decay of the exciton population follows due to recombination. In the coherent regime, by delaying the probe pulse by 100-fs steps, we are able to observe the effect of the pump-driven Rabi oscillations of the σ^+ density, resulting in the fast crossover between exciton and biexciton polaritons shown in Fig. 2a. When the pulses are temporally coincident ($\Delta t = 0$) we observe a practically pure biexciton-polariton doublet, while after 0.4 ps three polariton modes are present. After another 0.5 ps the field-driven Rabi oscillation has a maximum and the exciton polariton again disappears. Once the field-driven Rabi oscillations have completely died out, the density of incoherent σ^+ excitons is still high, and only the biexciton polaritons are detected.

In the incoherent regime a much slower crossover occurs (Fig. 2b): at a delay of approximately 25 ps the σ^+ population is depleted enough to let the σ^- excitonic transition reappear. After 100 ps the linear energies of the exciton polaritons are recovered in the $\sigma^+\sigma^-$ spectrum, and only a broadening persists. Therefore the phase-space filling phenomena (visible in the corresponding $\sigma^+\sigma^+$ spectrum at the same 100-fs delay) are negligible and the σ^- states are not relevantly populated by some spin flip mechanism.

The observed transfer of oscillator strength calls for a microscopic description. The theoretical approaches to the

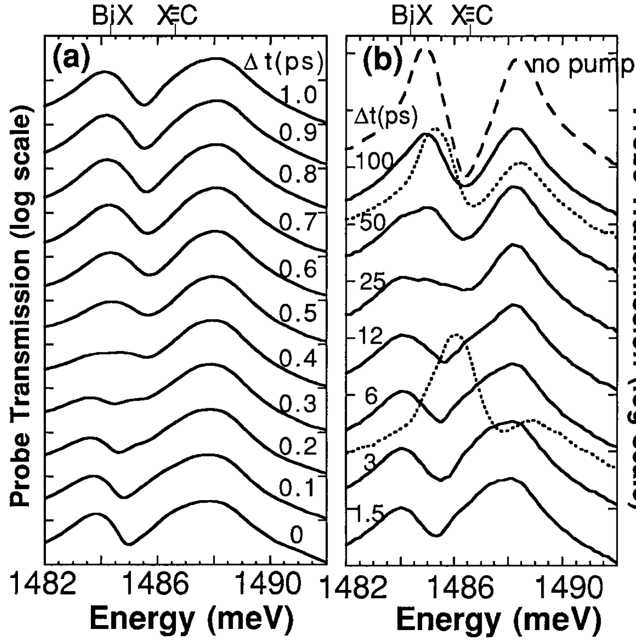

FIG. 2. Temporal dynamics of the crossover from biexciton to exciton polaritons: (a) coherent regime; (b) incoherent regime. The two dotted lines are the $\sigma^+\sigma^+$ spectra at 6 ps and 100 ps of delay (they are on the same scale as the corresponding $\sigma^+\sigma^-$). In both series the cavity energy is degenerate with that of exciton ($E_C = E_X = 1486.6$ meV) and the incident pump intensity is 44×10^{12} photons cm^{-2} pulse^{-1}.

biexcitonic correlation up to now have been mostly applied to the coherent response in the regime of low density ($\chi^{(3)}$ regime; see [8]). In the electron-hole basis, the correlation between exciton and biexciton polarization has been satisfactory described by means of four-particle terms. On the contrary, in the incoherent regime the pump polarization is lost and the correlation is given only by the real population, i.e., six-particle terms are required. A treatment of these terms in the electron-hole basis for 2D systems is not yet available, being numerically very demanding. Starting instead from the excitonic basis allows a direct interpretation of the correlation terms and an analytical solution of the equations, even if the features related to the free electron-hole pairs are neglected. This approximation is reliable in case excitons are generated resonantly and the creation of free carriers is limited. Our model starts from the Usui Hamiltonian for a gas of interacting excitons coupled to the external radiation field [16,17]. The Hamiltonian reads $H = H_0 + H_{\mathrm{field}} + H_{\mathrm{int}}$. The free term is $H_0 = \sum_{\sigma,\mathbf{k}} E_X(k) B^\dagger_{\sigma,\mathbf{k}} B_{\sigma,\mathbf{k}}$. The operator $B^\dagger_{\sigma,\mathbf{k}}$ creates an exciton with spin σ, wave vector $\mathbf{k}$, and energy $E_X(\mathbf{k})$. The coupling to an external radiation field $\Omega_\sigma(t)$ is accounted for by $H_{\mathrm{field}} = \sum_\sigma B^\dagger_{\sigma,\mathbf{0}}(1 - \frac{1}{An_{\mathrm{sat}}} B^\dagger_{\sigma,\mathbf{0}} B_{\sigma,\mathbf{0}})\Omega_\sigma(t)$ + H.c. Only excitons with $\mathbf{k} = 0$ are created (assuming that the wave vector of the radiation field is 0). The operators $B_{\sigma,\mathbf{k}}$ satisfy boson commutation rules, but the fermionic saturation of the exciton transition appears through the term inversely proportional to the density n_{sat} (A is the macroscopic quantization area). Finally, the Coulomb interaction between excitons is represented by the term $H_{\mathrm{int}} = \frac{1}{2}\sum_{\sigma,\sigma',\mathbf{k},\mathbf{k}',\mathbf{q}} V_q^{\sigma,\sigma'} B^\dagger_{\sigma,\mathbf{k}+\mathbf{q}} B^\dagger_{\sigma',\mathbf{k}'-\mathbf{q}} B_{\sigma,\mathbf{k}} B_{\sigma',\mathbf{k}'}$. Here, $V_q^{\sigma,\sigma'}$ is the spin-dependent interaction potential between excitons. In the following, we consider the response to the probe at "long" delays after the pump excitation, when coherence is lost. This allows an analytical solution for the probe susceptibility by calculating the one-exciton expectation value $p_-(t) = \langle B_{-,\mathbf{0}}\rangle$, that is, the polarization of the σ^- exciton mode with $\mathbf{k} = \mathbf{0}$. The probe being weak, all terms of order higher than 1 in the probe field are systematically neglected. We truncate the Coulomb correlation hierarchy to the third order, neglecting higher-order features (not observed in the experiment) and linearize the equations with respect to the exciton density. In the Heisenberg picture, the equation of motion for $p_-(t)$ is $\frac{\partial p_-(t)}{\partial t} = \frac{i}{\hbar}\{E_X(0)p_-(t) + \sum_{\mathbf{q},\mathbf{k}} V_q^{+,-} F_{\mathbf{k},\mathbf{q}}(t) + \Omega_-(t)\}$. Saturation does not occur in this equation, as only σ^+ population is present. The exciton transition amplitude $p_-(t)$, driven by the electric probe field $\Omega_-(t)$, is coupled through Coulomb interaction to the transition from the one-exciton to the biexciton state. This is formally expressed by the three-operator expectation value $F_{\mathbf{k},\mathbf{q}}(t) = \langle B^\dagger_{+,\mathbf{k}+\mathbf{q}} B_{+,\mathbf{k}} B_{-,\mathbf{q}}\rangle$, whose equation of motion is

$$\frac{\partial F_{\mathbf{k},\mathbf{q}}(t)}{\partial t} = \frac{i}{\hbar}\left\{[E_X(k) + E_X(q) - E_X(|\mathbf{k} + \mathbf{q}|)]F_{\mathbf{k},\mathbf{q}}(t) + \sum_{\mathbf{q}'} V^{+,-}_{|\mathbf{q}-\mathbf{q}'|} F_{\mathbf{k}-\mathbf{q}'+\mathbf{q},\mathbf{q}'}(t) + \delta_{\mathbf{q},\mathbf{0}}\Omega_-(t)N_{+,\mathbf{k}}\right\}. \quad (1)$$

The driving term of $F_{\mathbf{k},\mathbf{q}}(t)$ is proportional to the preexcited σ^+ exciton population $N_{+,\mathbf{k}} = \langle B^\dagger_{+,\mathbf{k}} B_{+,\mathbf{k}}\rangle$. Therefore the parameter $N_{+,\mathbf{k}}$ determines the relative oscillator strength of the two transitions. Notice that $F_{\mathbf{k},\mathbf{q}}(t)$ is coupled through exciton-exciton interaction to $F_{\mathbf{k}'',\mathbf{q}''}(t)$ such that $\mathbf{k} + \mathbf{q} = \mathbf{k}'' + \mathbf{q}''$. Since $N_{+,\mathbf{k}} = N_+\delta_{\mathbf{k},\mathbf{0}}$, only $F_{\mathbf{0},\mathbf{0}}$ has the source term $\propto \Omega_-(t)$. We can thus rewrite $F_{\mathbf{k},\mathbf{q}}(t) = \delta_{\mathbf{k}+\mathbf{q},\mathbf{0}}\Psi_{\mathbf{q}}(t)$. After this substitution the homogeneous part of Eq. (1) is just the Schrödinger equation for the biexciton problem, where $\Psi_{\mathbf{q}}(t)$ is the relative-motion wave function. We choose the rotating-wave frame and consider only the effect of the ground biexciton state: $\Psi_{\mathbf{q}}(t) = \beta e^{i\omega t}\Psi^{BIX}_{\mathbf{q}}$, where $\Psi^{BIX}_{\mathbf{q}}$ is the bound biexciton wave function in reciprocal space. Projecting Eq. (1) on the biexciton state [namely, operating with $\sum_{\mathbf{q}}(\Psi^{BIX}_{\mathbf{q}})^\star$], we are left with two linear coupled equations for p_- and β. The probe susceptibility $\chi(\omega,\xi) \propto \frac{p_-}{\Omega_-}$ is then obtained:

$$\chi(\omega,\xi) = \frac{f_X(1-\xi)}{\hbar\omega - E_X(0) - i\gamma} + \frac{f_X\xi}{\hbar\omega - [E_X(0) - b_{BIX}] - i\gamma},$$

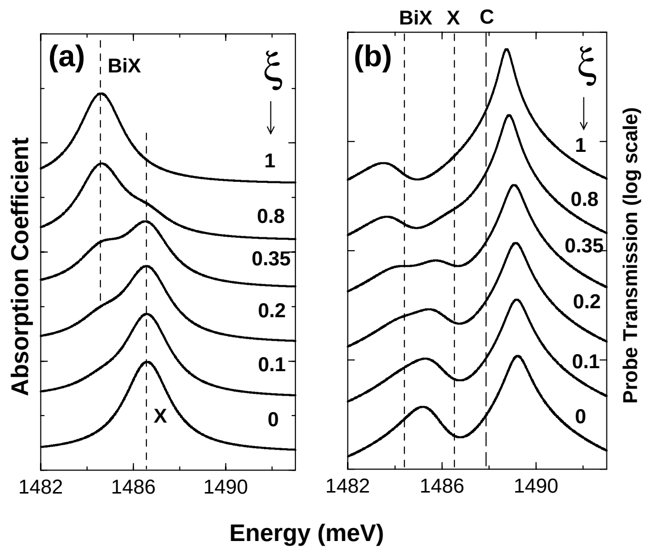

FIG. 3. (a) Calculated absorption spectra for the σ^- excitonic transition (proportional to the imaginary part of χ) for different values of the transfer factor ξ (defined in the text). (b) Corresponding transmission spectra obtained through a transfer-matrix calculation with the following parameters: $E_C - E_X = 1.3$ meV; $b_{BIX} = 2$ meV; $\gamma = 1.2$ meV.

where $\xi = |\Psi^{BIX}_{\mathbf{q=0}}|^2 N_+$, γ is a phenomenological broadening, b_{BIX} is the biexciton binding energy, and f_X is the exciton oscillator strength. The obtained susceptibility satisfies the sum rule $\int \hbar\, d\omega\, \chi(\omega, \xi) = 2\pi i f_X$ [18]. The transfer factor ξ has a clear physical interpretation. Taking a biexciton wave function $\Psi^{BIX}_{\mathbf{r}} \propto e^{-r/\lambda_{BIX}}$, one finds $\xi = n_+ 8\pi\lambda^2_{BIX}$, where n_+ is the total incoherent density of σ^+ excitons and λ_{BIX} is the biexciton radius. This means that the excitation of a σ^- exciton is inhibited in the biexcitonic area $8\pi\lambda^2_{BIX}$ surrounding a σ^+ exciton. The condition for the complete transfer of oscillator strength to the biexciton ($\xi = 1$) is obtained for an excited density $n_+ = n_{tr} = 1/(8\pi\lambda^2_{BIX})$.

The transmission spectra of the cavity are computed by inserting the optical susceptibility $\chi(\omega, \xi)$ of the QW in a transfer-matrix calculation (see Fig. 3). The agreement between theoretical and experimental spectra in the incoherent regime (Figs. 1c and 2b) is good for any value of ξ. As the coherent part of the polarization induced by the pump is not included in the model, the interpretation of the spectra in the coherent transient (Fig. 2a) remains instead qualitative.

Our theoretical model also permits us to estimate the ratio between the saturation density n_{sat} for the σ^+ transition and the transfer density n_{tr} observed in the $\sigma^+\sigma^-$ configuration. Within the Usui model one obtains $n_{\mathrm{sat}} = [\frac{16\pi}{7}\lambda^2_X]^{-1}$ [17], from which we find $n_{tr}/n_{\mathrm{sat}} = \frac{2}{7}(\lambda_X/\lambda_{BIX})^2 < 1$: the oscillator strength transfer takes place at a σ^+ exciton density that is lower than the saturation density for the σ^+ transition. This prediction can be experimentally verified: in Fig. 2b, 6 ps after the excitation, the Rabi splitting in the $\sigma^+\sigma^+$ spectrum is reduced with respect to the linear regime. The oscillator strength f of the σ^+ transition ($\propto\Delta^2$, the square of the Rabi splitting) can be estimated to be approximately 0.3 of the linear value f_X. At the same delay of 6 ps the $\sigma^+\sigma^-$ spectrum exibits a biexciton-polariton doublet. Thus, a full oscillator strength transfer occurs at an exciton density n_{tr} that is lower than the saturation density n_{sat}, as predicted by the model.

In conclusion, we report the experimental observation of an oscillator strength transfer from the exciton to the biexciton transition. The phenomenon is observed in a high quality microcavity by optically exciting a gas of σ^+ excitons and by probing the σ^- transmission spectrum. A sum rule holds stating that the total oscillator strength of exciton and biexciton transitions is independent of the density of σ^+ excitons. The transfer is theoretically described in terms of a three-exciton correlation induced by Coulomb interaction.

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