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Biexcitons or bipolaritons in a semiconductor microcavity

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A well-resolved nonlinear optical transition associated with biexcitons is observed in a high-quality microcavity with a Rabi splitting *exceeding* the binding energy of biexcitons in the embedded quantum well. This transition is identified as an induced absorption from the lower polariton to the biexciton. The biexciton binding energy is found not to be significantly affected by the coupling with the cavity photons. In spite of the high quality of the sample, formation of a bipolariton as a bound state of two lower cavity polaritons with small in-plane wave vectors is not resolved.

The formation of biexcitons in semiconductors has attracted considerable attention in recent years.[1] Especially intriguing is, from the fundamental point of view, the problem of a biexciton in the presence of two types of interactions: the exciton-exciton attraction by the Coulomb interaction between the constituent electrons and holes, leading to the biexciton formation, and the exciton-photon coupling which leads to the formation of polaritons.[2] The possibility to realize low-dimensional heterostructures has introduced a further variable to the problem. In low-dimensional systems, such as quantum wells (QW's), the Coulomb interaction is enhanced by the spatial confinement. The interaction of excitons with light is also strongly modified by the confinement. In two-dimensional (2D) systems, the breaking of translational invariance along the growth direction leads to the coupling to a continuum of photon modes, resulting in an irreversible radiative decay of the excitons, while in the bulk case the polariton is a stationary state.[2] The influence of the exciton-photon coupling on the internal structure of biexcitons has been discussed in 3D, 2D, and 1D systems.[3]

The recent development of microcavity structures has opened new possibilities to control the light-matter interaction.[4,5] A quantum-well microcavity is a planar Fabry-Perot cavity with an optical length of an integer multiple of half-wavelengths of the photons resonant to the QW excitons embedded in the cavity. Photon quantization along the cavity axis occurs, and excitons with a given in-plane wave vector couple to only one photon mode. Stationary eigenstates of the system now exist, called cavity polaritons, and an irreversible radiative decay of excitons no longer occurs. In real systems the photons in the cavity have a finite lifetime due to transmission through the mirrors. Thus, cavity polaritons are converted into external photons as a result of the finite lifetime of their photonic component in the cavity. The strong exciton-photon coupling that occurs in a microcavity significantly modifies the optical properties of the embedded QW excitons. When the vacuum Rabi splitting in the microcavity, which quantifies the strength of the exciton-photon coupling, is significantly bigger than the biexciton binding energy one would imagine that the optical properties of biexcitons are also affected by the coupling with the cavity photons. In particular, the formation of a bound two-polariton system (bipolariton) could be expected.

The experimental observation of biexcitons in microcavity was addressed recently by Fan *et al.*[6] and by Neukirch *et al.*[7] Fan *et al.* could not distinguish a well-resolved biexciton transition due to a large broadening of the investigated polariton resonances. Alternatively, Neukirch *et al.* used a nonmonolithic II-VI microcavity with a biexciton binding energy that exceeds the vacuum Rabi splitting and all damping constants. This structure allowed them to spectrally separate biexcitonic contributions from the polariton resonances. However, such a structure is not suitable to investigate the effect of a strong exciton-photon coupling on the binding of biexcitons in microcavities, which was, thus, not addressed in their work. They measured coherent spectral oscillations in a pump-probe experiment at *negative* pump-probe delay times which included coherent features related to an upper polariton-biexciton transition. However, an incoherent pump-induced absorption associated with this transition at *positive* delay times was not observed.

In this paper we investigate the nonlinear optical properties of a very high-quality monolithic QW microcavity in a pump-probe experiment. The heavy-hole vacuum Rabi splitting in the microcavity is approximately four times bigger than the biexciton binding energy in the bare QW. Due to the narrow linewidth of the polariton resonances, a well-resolved pump-induced optical absorption associated with biexcitons is observed. The energy position, the dependence on the polarization of pump and probe photons, and the dependence on the detuning between the cavity mode and the exciton clearly demonstrate the biexcitonic nature of this resonance. The biexciton binding energy is found not to be significantly affected by the strong exciton-photon coupling in the microcavity.

The sample consists of a molecular-beam epitaxy–grown 25 nm $GaAs/Al_{0.3}Ga_{0.7}As$ single quantum well placed in the center of a λ cavity.[8] An $AlAs/Al_{0.15}Ga_{0.85}As$ Bragg reflector of 25 (16) periods was grown at the bottom (top) of the cavity. The $Al_{0.3}Ga_{0.7}As$ barriers of the QW are 5 nm thick and the rest of the spacer layer consists of $Al_{0.05}Ga_{0.95}As$. The spacer layer is wedged, in order to tune the cavity mode

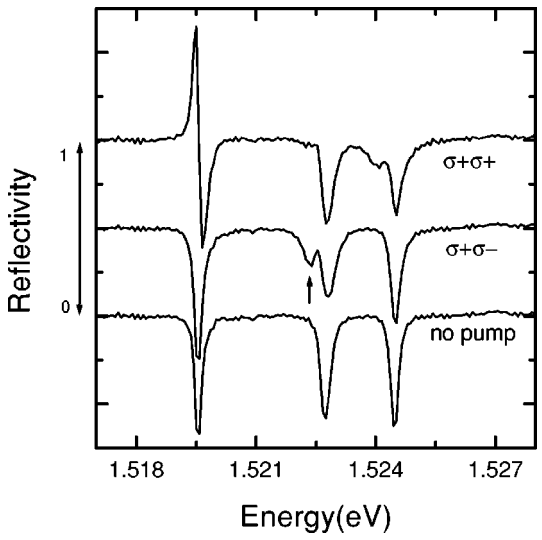

FIG. 1. Reflectivity spectra for co-circular and cross-circular pump and probe photons at zero pump-probe delay. The cavity mode is in resonance with the bare HH exciton (zero detuning). The pump fluence is 0.12 μJ/cm^2. The lower curve is the reflectivity spectrum without the pump pulse, shown for reference.

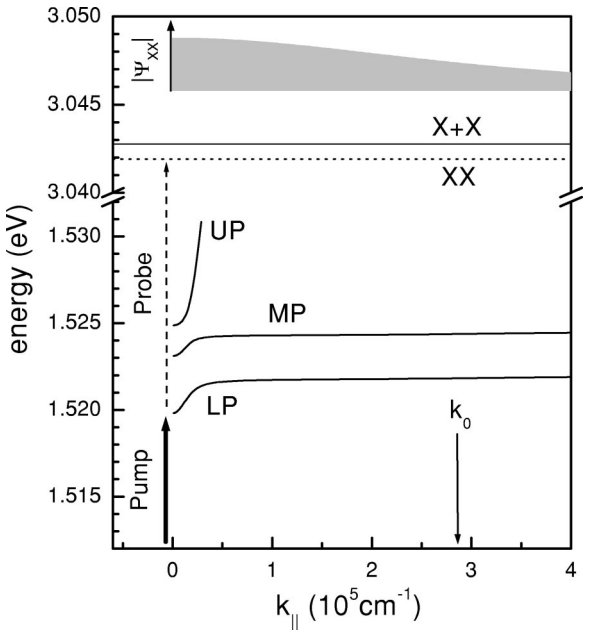

FIG. 2. Lower (LP), middle (MP), and upper polariton (UP) dispersion versus the in-plane wave vector. The radiative cutoff k_0 is indicated. The biexciton (XX) and twice the exciton energies ($X+X$) are also shown. The amplitude of the biexciton wave function of the exciton-exciton relative motion extending to large wave vectors of one of the constituent excitons, comparable to the radiative cutoff, is depicted in gray scale. The absorption of pump photons (solid arrow) creates a LP density that induces a LP-XX absorption of probe photons (dashed arrow).

along the position on the sample. Details on the growth conditions and sample design can be found in Ref. 9. The pump-probe experiment has been performed using a Ti:sapphire laser source providing Fourier-limited 100 fs laser pulses at 76 MHz repetition rate. The spectrum of the pump pulse was shaped in order to resonantly excite only the lower polariton in the microcavity, resulting in a pulse duration of $\sim$500 fs. Small incidence angles ($\sim 1°$) for both pump and probe were used and the probe reflectivity was spectrally analyzed with a spectrometer and a charge-coupled device camera of 70 μeV resolution (half width at half maximum-HWHM). The probe spot size was $\sim$40 μm, resulting in a small energy broadening of the cavity mode from the wedge gradient and the incident wave-vector spreading.[9] The pump spot size was chosen 40% bigger than the probe, in order to have a uniform pump density over the probe area. The sample was held in a helium bath cryostat at $T=5$ K for all the measurements. A variable delay time between pump and probe pulses and Babinet-Soleil plates for polarization control were used.

The probe reflectivity spectrum without the pump pulse is shown in Fig. 1 with the cavity mode in resonance with the bare heavy-hole (HH) quantum-well exciton (zero detuning). The wide GaAs quantum well has a narrow and homogeneously broadened bare QW exciton absorption resonance.[10] The strong exciton-photon coupling in the microcavity structure leads to three polariton resonances, arising from the mixing of the heavy-hole exciton, the light-hole (LH) exciton and the cavity mode. Measurements at 11 K of the polariton resonances for different detuning between the cavity mode and the HH exciton were fitted by a three coupled-oscillator model from which we have inferred a Rabi splitting of 3.6 meV for the HH and 2.2 meV for the LH excitons.[9] Moreover, the use of a high resolution spectrometer allowed us to accurately measure the polariton linewidth, which was found to be 95 μeV HWHM at each of the resonances at zero detuning, resulting in a ratio of 19 between the HH Rabi splitting and the linewidth.[11] The probe reflectivity in presence of the pump pulse at zero relative delay is shown in Fig. 1 for two circular polarization combinations of pump and probe as indicated. The pump pulse leads to a well distinguished absorption resonance slightly below the middle polariton (MP) that disappears for co-circular polarizations (see arrow in figure).[12] The formation of heavy-hole biexcitons in a bare QW is known to follow these polarization selection rules, namely, is canceled for co-circular excitation.[10]

In a recent theoretical work,[13] the main difference between the lowest bipolariton bound state in a microcavity, formed by two lower polaritons (LP) with small in-plane wave vectors ($\mathbf{k}_\parallel$), and the bare biexciton has been pointed out. The total energy of the bipolariton is twice the LP transition energy minus the bipolariton binding. Thus, by populating the LP branch at small $\mathbf{k}_\parallel$ with the pump pulse, an induced absorption from the LP to the bipolariton should lead to a reflectivity dip *below* the LP, separated by the bipolariton binding. However, due to the small effective mass in the LP, the bipolariton binding energy is very small compared to the bare biexciton value. It scales exponentially with the mass ratio between LP and exciton and is negligible compared to the unavoidable damping of the LP,[13] even at very low temperatures due to the radiative damping. Thus, a bipolariton resonance is not expected to be resolved in microcavities. Instead, the bare biexciton radius, typically comparable to the exciton Bohr radius, corresponds to a biexciton wave function that extends to large in-plane wave vectors of the constituent excitons ($\mathbf{k}_\parallel \sim 2\pi n/\lambda$). Therefore, even inside the microcavity, the biexciton bound-state structure

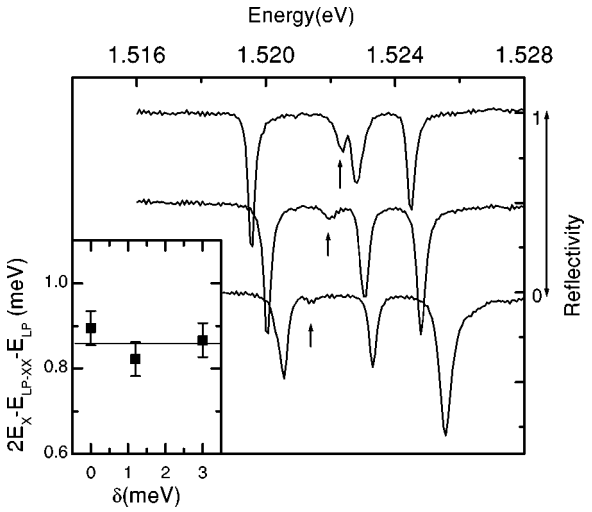

FIG. 3. Reflectivity spectra at zero pump-probe delay for different detuning between the cavity mode and the HH exciton. The pump fluence is 0.12 μJ/cm^2. The inset shows the difference between twice the HH bare exciton energy and the measured sum of the LP and LP-XX transition energies. The line is a guide for the eyes. The detuning δ is defined as the energy difference between the cavity mode and the bare HH exciton.

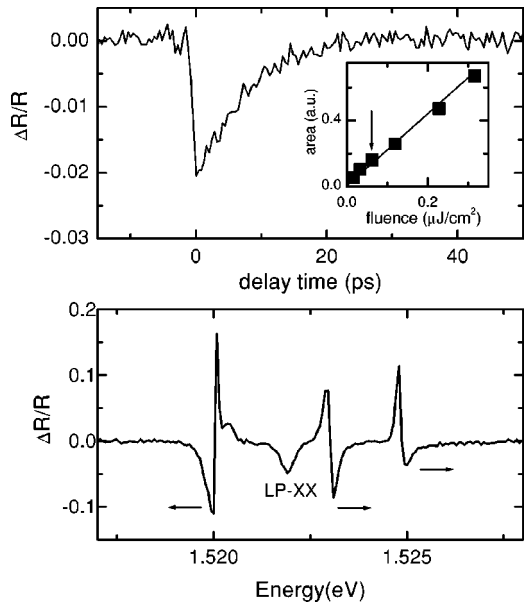

FIG. 4. Upper plot: Differential reflectivity time-resolved and spectrally integrated over the LP-XX transition. Inset: time-integrated differential reflectivity versus pump fluence. The arrow indicates the fluence used in the curve shown. Lower plot: Differential reflectivity spectrum at zero delay. The arrows indicate the shifts of the LP, MP, and UP that occur in presence of the pump pulse. All data are taken with the cavity mode 1.2 meV above the bare HH exciton (positive detuning).

(i.e., binding energy and radius) is not expected to be significantly modified by the photon coupling which is affecting only the dispersion region at much smaller $\mathbf{k}_\parallel$.[13] The transition from the LP to the biexciton is *higher* in energy than the LP transition,[14] by the amount: $E_{\mathrm{LP}-XX}-E_{\mathrm{LP}}=2(E_X-E_{\mathrm{LP}})-E_{BXX}$, with E_{LP}, E_X, $E_{\mathrm{LP}\text{-}XX}$ being the lower polariton, bare exciton, and LP-biexciton transition energies, respectively, and E_{BXX} the biexciton binding energy. These concepts are sketched in Fig. 2. The dispersion of the LP, MP, and upper polariton (UP) branches are calculated for the investigated structure at zero detuning. The radiative cutoff of the exciton-photon coupling $k_0=2\pi n/\lambda$ is indicated. The biexciton energy (XX) and twice the bare exciton energy ($X+X$) are indicated, the difference between them being the biexciton binding energy. The pump photons create a LP density at $\mathbf{k}_\parallel\sim 0$. When the probe arrives, an induced absorption from the LP to the biexciton transition occurs. The amplitude of the optically active biexciton wave function with the wave vector $K\sim 0$ of the in-plane center of mass motion is also shown versus the in-plane wave vector of one of the constituent excitons. This wave function describes the exciton-exciton relative motion with a relative wave vector, at $K=0$, which is twice the one of the constituent exciton. We have calculated this wave function using the 2D deuterium ground-state formula with the biexciton radius given by $1/\sqrt{8}k_0$ that follows from equal radiative rates of the biexciton and the exciton,[1] as experimentally measured in a bare QW.[10] The biexciton wave function extends to large wave vectors, even above the radiative cutoff.

That the observed induced absorption shown in Fig. 1 corresponds to the LP-XX transition is further confirmed by changing the detuning between the cavity mode and the HH exciton. When shifting the cavity mode above the HH exciton (positive detuning) the LP transition shifts towards higher energies.[9] Since the XX energy is not significantly affected by the coupling to the cavity photons and therefore by the detuning, the LP-XX transition should shift towards lower energies. This is shown in Fig. 3 for three different detunings. The LP-XX transition is indicated by the arrows, and clearly shifts towards lower energies while the LP shifts towards higher energies. This also results in a reduced visibility of the LP-XX resonance while it shifts far from the cavity resonance. The expected shift of the LP-XX transition versus the detuning is given by $E_{\mathrm{LP}\text{-}XX}=2E_X-E_{BXX}-E_{\mathrm{LP}}$ with the detuning dependent E_{LP}. Thus, the sum of $E_{\mathrm{LP}\text{-}XX}$ and E_{LP} is expected to be independent of the detuning. In the inset of Fig. 3 the difference between twice the bare HH exciton energy and the measured sum of LP and LP-XX transition energies is shown versus the detuning. Within the experimental uncertainty, a constant biexciton binding energy of 0.86 meV is obtained. This value is slightly smaller than the 1.1 meV measured in a comparable bare QW.[10] We attribute the reduction to an increased dielectric screening of the Coulomb attraction, as a consequence of the higher refractive index of the QW barrier in the microcavity structure compared to the bare QW.[9,10] Additionally, built-in electric fields would also reduce the biexciton binding. The presence of excess free carriers, that could lead to such electric fields, was in fact observed in the investigated structure.[11] A constant value of E_{BXX}, independent of the detuning, shows the negligible influence of the exciton-photon coupling to the biexciton binding.

The dynamics of the LP-XX induced absorption is ruled by the LP lifetime. When the LP density created by the pump is fully decayed into external photons via the cavity photon lifetime, the LP-XX induced transition disappears. This is, in fact, observed by measuring the induced absorption as a function of the pump-probe delay (see Fig. 4). Positive delay

corresponds to the pump leading the probe pulse. In the top part of Fig. 4 is shown the differential reflectivity at the LP-*XX* transition, spectrally integrated over the resonance. The lifetime of the induced absorption is well fitted by an exponential decay with a time constant of 6.5 ± 0.5 ps. The cavity photon lifetime in the investigated structure is 2.2 ps, as deduced by the cavity linewidth of Ref. 9 ~ 300 μeV. At the detuning corresponding to the data shown in Fig. 4 we calculated a photonic content of 0.37 in the LP that results in a LP radiative lifetime of 6 ps, in agreement with the fit within errors. In the inset is shown the dependence of the time-integrated differential reflectivity at the LP-*XX* transition on the pump excitation intensity. A linear dependence is observed, as expected from a LP-*XX* absorption proportional to the pump-induced LP density.

Finally, the lower part of Fig. 4 shows the spectrally resolved differential reflectivity at zero delay. Compared to the direct reflectivity data, the LP-*XX* transition is resolved with better signal-to-noise ratio in the differential reflectivity spectrum. The LP-*XX* energy positions used in the inset of Fig. 3 were measured from the differential reflectivity spectra to reduce the uncertainty. An additional feature is observed in the spectrum. In the presence of the pump pulse the LP, MP, and UP energies are slightly shifted (shifts are below 100 μeV) as indicated by the arrows, corresponding to an increase of the HH rabi splitting. This was observed at all investigated pump intensities at 1.2 meV and 3 meV detunings, but less pronounced at zero detuning. Similar observations have been reported,[6] which were, however, based on experimental findings dominated by the inhomogeneous broadening in the microcavity resonances, and attributed to the induced biexciton transition within a phenomenological model. Based on the high quality of the presented data, further theoretical work for a detailed understanding of this effect is strongly encouraged.

In conclusion, we have experimentally shown the presence of a well-resolved nonlinear optical transition associated with biexcitons in a quantum-well microcavity of high quality in the strong coupling regime. This transition is identified as an induced absorption from the lower polariton to the biexciton. Even if the vacuum Rabi splitting in the microcavity is four times bigger than the biexciton binding energy in the bare quantum well, the biexciton binding energy is not significantly affected by the coupling with the cavity photons. In contrast, the formation of a bipolariton as a bound state of two lower cavity polaritons with small in-plane wave vectors is not resolved. These results are in agreement with theoretical expectations in literature based on the in-plane wave-vector dispersion in microcavities.[13,14]

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