Saturation of the strong-coupling regime in a semiconductor microcavity: Free-carrier bleaching of cavity polaritons

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We present experimental data on saturation of the strong-coupling regime in semiconductor microcavities based on intensity-dependent photoluminescence measurements. The saturation can be understood in terms of electron-hole pair screening of the quantum-well exciton. The very low saturation intensity $I_{\rm sat}^{\rm incident}=100\ {\rm W\,cm^{-2}}$ leads to a saturation density $N_{\rm sat}^{100\ {\rm K}}=4.3\times10^{10}\ {\rm cm^{-2}}$ in good agreement with a theoretical model. These results are important for applications such as lasers in the strong-coupling regime and nonlinear devices.

There is increasing interest in the physics and potential application of the strong-coupling regime in semiconductor microcavities.[1–6] This regime occurs when an excitonic state of a quantum well (QW) is brought into resonance with a discrete Fabry-Pérot mode (FP) of a microcavity, and when the Rabi frequency of the coupled system is larger than any dephasing time or than the lifetime of both oscillators. In other words, the interaction energy must be larger than any homogeneous or inhomogeneous broadening of the uncoupled modes or energy levels. In the strong-coupling regime, normal-mode splitting occurs which lifts the degeneracy of the excitonic and photon oscillators. The splitting Ω is a function of the oscillator strength ($f_{\rm osc.}$), the number of quantum wells ($N_{\rm QW}$), and the cavity length ($L_{\rm cavity}$):

$$\Omega\propto\left[\frac{f_{\rm osc.}N_{\rm QW}}{L_{\rm cavity}}\right]^{1/2},$$

or, if finite oscillator linewidths are considered,

$$\Omega\propto\left[\frac{(f_{\rm osc.}-f_{\rm th.})N_{\rm QW}}{L_{\rm cavity}}\right]^{1/2},$$

where $f_{\rm th.}$ is a threshold oscillator strength whose value depends on the exciton and FP linewidths. The decay of the normal mode depends in a nontrivial way on the lifetimes of the uncoupled exciton and FP oscillators, and in an ideal system the coupled exciton-photon state would not decay. This is in contrast with the usual weak-coupling regime: an electronic state, being coupled to a continuum of photon final states, irreversibly decays into the continuum before a Rabi oscillation can be accomplished. In this regime, a perturbative approach like Fermi's golden rule is a valid description of the light-matter interaction, and the coupled and uncoupled eigenstates are essentially the same. This is no longer true in the strong-coupling regime.

It has been suggested that the strong-coupling regime could have an important impact on optoelectronic devices.[7] This point is of particular interest since the strong-coupling regime has been observed up to room temperature.[2] Additional types of lasers and low-noise light sources have been proposed as well as electrooptic modulators, and absorptive elements. Up to now, most studies of the strong-coupling regime dealt with its basic physical properties, but there has been little attention given to applications and devices.

Saturation of the light-matter interaction as a function of the light intensity or as a function of the electronic state density is a well-known effect in semiconductors, and is the basis of many important applications. Such effects are of great importance to complete our understanding of the strong-coupling regime, and for applications involving high light intensities or high carrier densities. In fact the strong-coupling regime is a very good tool for studying saturation effects: the simple relation between the mode splitting and the exciton oscillator strength enables a *direct* and precise measure of $f_{\text{osc.}}$. Moreover, because the band-gap renormalization just compensates for the decrease of the exciton binding energy,[8] *the resonance condition is always fulfilled during the bleaching process.* Only at a much higher excitation intensity does the FP mode become degenerate with the electron-hole pair continuum, whose gap lies at or below the FP mode energy. This forms an important system, where a continuous transition from a discrete strongly coupled state to a weakly coupled continuum can be observed. In this paper we report on the saturation of the strong-coupling regime by nonresonantly excited electron-hole pairs.

The sample consists of a $3\lambda/2$-long GaAs cavity with six $In_yGa_{1-y}As$ QW and two $AlAs/Al_{0.1}Ga_{0.9}As$ Bragg mirrors. It is described in more detail elsewhere.[2,3] By design, the cavity is wedge shaped, leading to a variation of the relative position of the QW exciton and the cavity mode across the sample. The sample is excited with a Ti sapphire. The laser beam is chopped by an acousto-optic modulator to avoid sample heating. In order to obtain a homogeneous excitation power density, the light is then coupled into a 5-m-long multimode fiber (100-μm diameter). The fiber output is then imaged on the sample with a 2:1 magnification. The sample is mounted on a cold finger at a temperature of 100–110 K. An angular emission analysis is achieved by rotating the collection optics around the excited spot on the sample. The aperture of the collection cone is 5°. Due to restrictions on the sample alignment, the angular accuracy is only ±2°. This uncertainty was not quoted in Ref. 3.[9] All experiments are performed under *nonresonant* excitation at an energy between the GaAs and $Al_{0.1}Ga_{0.9}As$ band gaps; i.e., electron-hole pairs are initially created inside the whole GaAs cavity.

Saturation studies of strong coupling may be done either in absorption or photoluminescence (PL). PL was chosen for experimental convenience. In the strong-coupling regime two transitions, which are a direct signature of the two normal modes, are observed,[10,11,2,3] while in the weak-coupling regime, *at resonance,* only one transition, modified by the FP cavity (acting as a filter) is observed. In the strong-coupling regime it has been shown[12] that absorption and PL are simply connected by a Boltzmann thermal distribution function. Weak and strong couplings are also differentiated by a crossing or an anticrossing behavior of the eigenstate energies near the resonance condition. This can be observed as a function of the detuning[1] or in the dispersion curves in reciprocal space.[3] The last point leads to the concept of the cavity polariton. We have recently demonstrated that angle-resolved photoluminescence (PL) experiments allow the direct measurement of the cavity-polariton dispersion curve.[3]

Two spectra taken at low (a) and high (b) excitation power densities are shown in Fig. 1. The low-intensity spectrum exhibits the doublet structure characteristic of the strong-coupling regime. The excited spot on the sample is selected to have exact resonance between the exciton QW and the FP mode at normal incidence. Both lines have different relative intensities because of different Boltzmann population factors. The high-intensity spectrum exhibits only one single broad line characteristic of a weak-coupling regime. Note the symmetric splitting of the strongly coupled lines (a) with respect to the uncou-

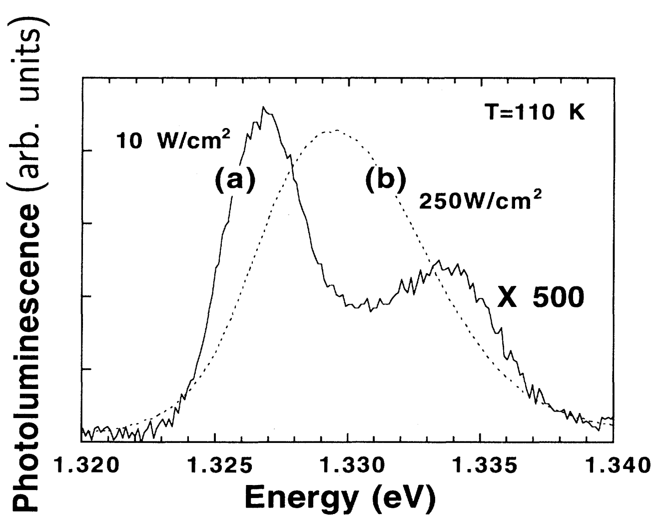

FIG. 1. 110-K cavity-polariton photoluminescence spectrum under low [(a), continuous line] and high [(b), dashed line] excitation power density under nonresonant excitation.

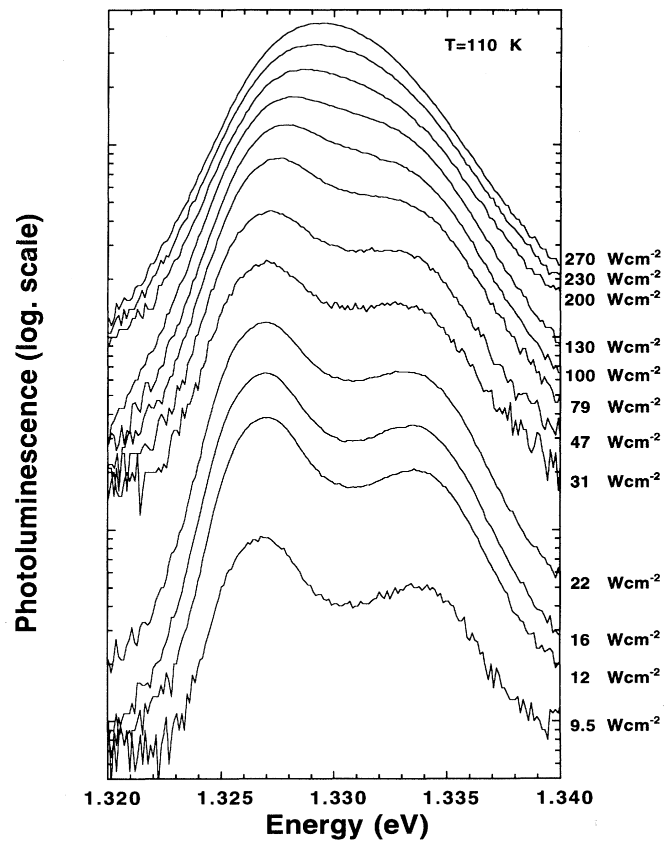

FIG. 2. Series of photoluminescence spectra (110 K) as a function of pump power showing bleaching of the strong-coupling regime. The incident power density is shown on the right axis.

pled energy level as measured from the weakly coupled spectrum (b). This confirms the zero detuning between the exciton and FP modes. A series of spectra as a function of the incident pump power is shown Fig. 2. The bleaching is clearly observed, although the doublet is not as well defined because of the logarithmic scale.

A good proof that what occurs is a decrease of the interaction energy (i.e., the oscillator strength) rather than a shift of an oscillator away from the resonance condition is that the anticrossing behavior, characteristic of the strong-coupling regime, turns into a crossing behavior at high intensity. These differences are clearly seen in angle-resolved PL measurements which were performed under low and high excitation densities. Figure 3(a) shows such a measurement taken at a point on the sample, where the resonance condition is fulfilled at a finite emission angle. The anticrossing between the exciton and FP dispersion curves is clearly observable. For more details on the interpretation of this type of measurements, the reader should refer to Ref. 3. On the other hand, under high excitation [Fig. 3(b)], the cavity polariton is bleached, and the exciton and FP dispersion curves now cross each other; i.e., both oscillators are weakly coupled. Note that the relative intensity of the PL lines has changed, indicating that the dynamics have also changed. This is especially clear near normal incidence.

Oscillator strength as a function of the electron-hole density was extracted from the data of Fig. 2, using a transfer-matrix formalism[13] to calculate absorption spectrum and assuming a Boltzmann distribution along the dispersion curve. The following procedure was used: linewidths of both uncoupled oscillators γ_X and γ_{FP} were measured from a PL spectrum taken far off resonance. As γ_{FP} is usually found to be larger than the theoretical empty cavity value, losses in the cavity are included to fit the measured value of γ_{FP}. This last point enables much better agreements than previously reported.[2] The oscillator strength remains the last free parameter, and is used to fit the splitting Ω. No significant broadening was observed experimentally up to the complete bleaching of the strong-coupling regime. From lifetime measurements,[14] $\tau \approx 1.5$ ns in the range of excitation power intensity used in this study. The calculated absorption of the pump light is 45% in the GaAs cavity. The plot of the oscillator strength vs electron-hole density is shown in Fig. 4, assuming that all the excited carriers are evenly shared between the six QW's in the cavity. This last assumption will tend to overestimate the electron-hole pair density, as the capture efficiency is probably less than 100%.[15] The measurements can be well fitted by a usual screening function:[16] $f(N_{e\text{-}h}) = f_0/(1+N_{e\text{-}h}/N_{sat})$ with $N_{sat} = 4.3\times10^{10}$ cm^{-2}. This corresponds to a remarkably low incident saturation intensity $I_{sat}^{incident} = 100$ W cm^{-2} of the cavity polariton. The value of N_{sat} is significantly lower than the one reported by Chemla and coworkers[16,17] for $Al_xGa_{1-x}As$/GaAs QW's at room temperature ($N_{sat}^{300\,K} = 5\times10^{11}$ cm^{-2}). Nevertheless, considering the much larger exciton Bohr radius a_B of

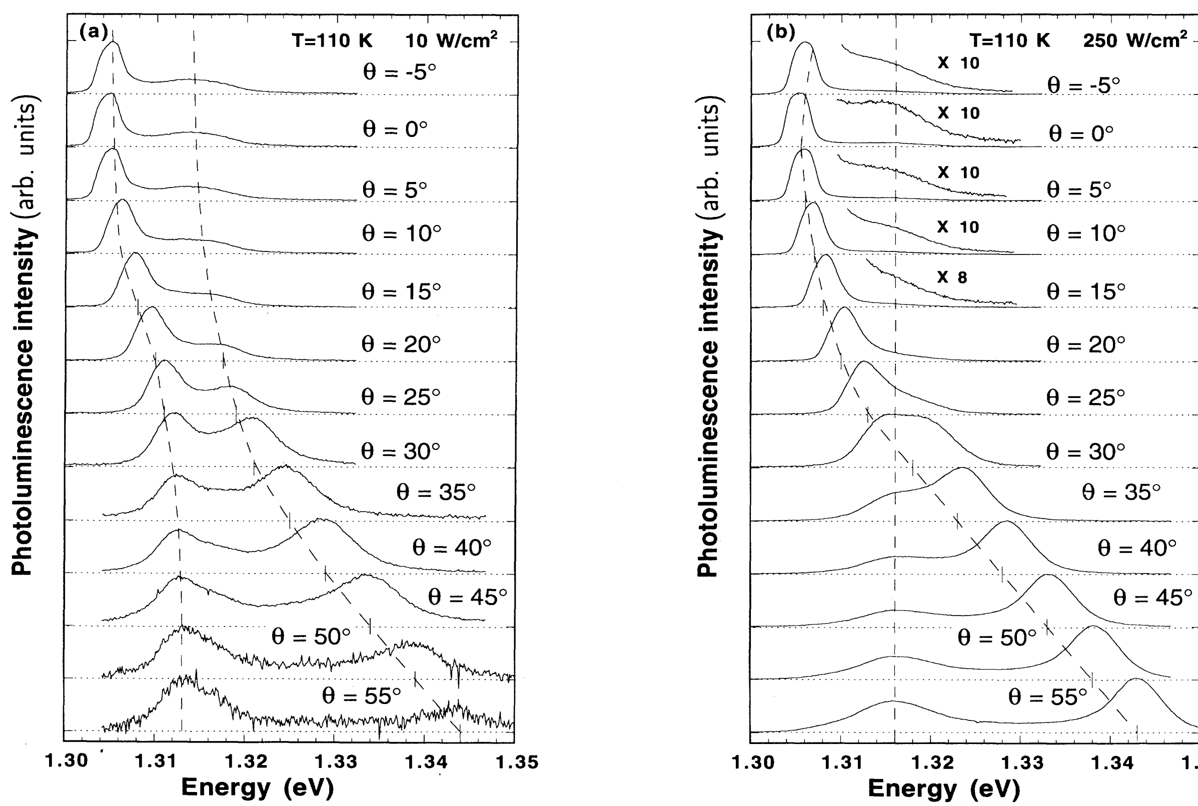

FIG. 3. Angle-resolved photoluminescence under (a) low and (b) high excitation. Dashed lines are a guide to the eye to follow the respective anticrossing and crossing behaviors of the dispersion curves. For more details on the interpretation of this type of measurement, the reader should refer to R. Houdré *et al.*, Phys. Rev. Lett. **73**, 2043 (1994).

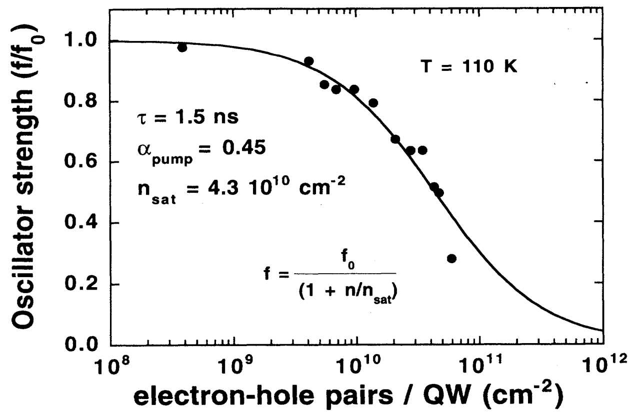

FIG. 4. Exciton oscillator strength as a function of electron-hole pair density. The continuous line is a fit with a usual screening function (Ref. 16) $f(N_{e\text{-}e})=f_0/(1+N_{e\text{-}h}/N_{sat})$.

$In_yGa_{1-y}As/GaAs$ QW's [$a_B=120$ (Ref. 18)–140 (Ref. 19) Å], and the more efficient bleaching by electron-hole pairs at lower temperature, the same model of Schmitt-Rink, Chemla, and Miller[17] gives $N_{sat}^{100\,K}=2.1\times10^{10}$ cm^{-2} with $a_B=140$ Å (which separates into 5.4×10^{10} cm^{-2} due to phase-space filling and 3.5×10^{10} cm^{-2} due to exchange screening), in good agreement with the experimental value.

To comment on the excitation intensity dependence of the PL spectra in the weak-coupling regime is a much more delicate task, because $f_{osc.}$ is no longer directly measured from the spectra, and the relation between PL and absorption is no longer a thermal factor. Such comments will not be attempted in this paper; nevertheless it can be noted that the linewidth at higher intensity is consistent with an optical transition with the band-to-band continuum. Assuming 1.5% absorption per QW (Ref. 20) gives a linewidth of 9 meV as compared to the 7.5–8-meV experimental value. An excitonic transition in the weak-coupling regime would have given $\gamma_{PL}\approx\gamma_{FP}\approx1$ meV.

These results are also important for device applications. Lasers in the strong-coupling regime have been proposed as an approach to the so-called thresholdless lasers.[7] From its very definition, as the excitonic state in the strong-coupling regime is only coupled to a single-photon mode and emission or coupling into other photon modes can be neglected, we are in exactly the $\beta\approx1$ condition.[21] This condition can easily be achieved under resonant optical pumping,[22] where only coupled exciton and photon states are created. The issue of electrical or nonresonant pumping has not yet been addressed. The present result demonstrates that additional concepts have to be explored in order to reach lasing action before destroying the strong-coupling regime for nonresonantly pumped structures.

For applications such as bistability, low saturation density and consequently low saturation intensity are of great interest. One could think of an absorptive bistability based on a switching mechanism between strong- and weak-coupling regimes induced by a bleaching of the exciton oscillator strength: The bleaching of $f_{osc.}$ decreases the normal-mode splitting, possibly leading to positive feedback on the absorption in the energy range $[E_X-\Omega/2,E_X+\Omega/2]$. Such applications are beyond the scope of this paper and will be commented in a further article.

In conclusion, we have presented experimental data on saturation of the strong-coupling regime in semiconductor microcavities. The saturation of the QW exciton by free-carrier screening explains the observation. The saturation density $N_{sat}=4.3\times10^{10}$ cm^{-2} is in good agreement with the theoretical model, and leads to a very low saturation intensity $I_{sat}^{incident}=100$ W cm^{-2}. These results place fundamental limitations on certain applications such as lasers in the strong-coupling regime, and are important for nonlinear devices.

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