

Measurement of Cavity-Polariton Dispersion Curve from Angle-Resolved Photoluminescence Experiments

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We study the photoluminescence of quantum well excitons imbedded in monolithic microcavities. In the strong coupling regime, a coupled-mode situation develops, the cavity polariton. We describe a model of the photoluminescence phenomenon in this regime, which by comparison with experiments enables us to determine the cavity-polariton dispersion curve. An excellent agreement with a theoretical model is found.

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Since the early studies of Lukosz and Kunz [1], Drexhage [2], and Kleppner [3], great attention has been devoted to the study of the modification of the photon-matter interaction due to quantized electromagnetic modes near surfaces [1,2], in microcavities [4–7], and in photonic band-gap materials [8], the matter being in various forms like atoms, dye molecules, semiconductors, etc. One aim is to control the spontaneous emission in its intensity and emission pattern [4], in its spectral properties [5,6], and in its fluctuations [7] (i.e., generation of squeezed photon states). Another aspect of fundamental interest is the possibility of generating well-defined atom-photon quantum states or nonlocal field states, the so-called Schrödinger cats [9].

The photon-matter interaction has two distinct regimes: (i) the weak-coupling regime, where the system's eigenstates are a perturbation of the uncoupled system; and (ii) the strong-coupling regime, where coherent Rabi oscillation between the atom and photon states can occur. Both have been studied and lead to very different and interesting situations. The strong coupling existing between photons and quantum well (QW) excitons imbedded in all-semiconductor planar microcavities has been recently observed, first at low temperatures, and more recently at room temperature [10,11]. The physical picture is the following: A QW exciton state, with a well-defined transverse k momentum (k_{\perp}), is coupled to a single cavity-photon mode with the same in-plane wave vector, due to momentum conservation caused by the in-plane translational invariance of the electronic and photon systems. This selection mechanism explains why the single atomic picture of two coupled levels applies here, although we are dealing with a pair of two-dimensional continuum systems. For a given k , a single photon mode and the *single* coupled exciton interact with an equivalent oscillator strength of about 10^{12} atoms [12]. Provided that the exciton momentum scattering time (dephasing time) and the photon-mode decay time (photon lifetime) are longer than the Rabi period, an oscillating regime between the exciton and photon state develops, the very definition of

the strong-coupling regime. It must be remarked that this phenomenon can occur not only under strong applied optical fields, but also when no field is applied, leading to the so-called vacuum field Rabi splitting in atomic physics [13] and to cavity polaritons in semiconductor microcavities [11]. Several descriptions of this phenomenon have been provided, based either on the classical dispersion model [14,15] of vacuum field Rabi splitting or more recently on the quantum description of exciton and photon modes [16,17]. The splitting Ω is a function of the electric dipole matrix element of the atomic transition (d), the number of atomic oscillators (N_{at}), or, in semiconductors, a function of the oscillator strength (f_{osc}) and the number of quantum wells (N_{QW}) and the cavity size (ν_{cavity}) (i.e., the length for a planar cavity or the volume for a 3D cavity):

$$\Omega \propto d \sqrt{\frac{N_{\text{at}}}{\nu_{\text{cavity}}}} \propto \sqrt{\frac{f_{\text{osc}} N_{\text{QW}}}{\nu_{\text{cavity}}}}$$

As remarked elsewhere [10], this situation is very reminiscent of the strong-coupling regime observed about thirty years ago in bulk semiconductors, leading to excitonic polaritons [18]. In that case, it was remarked by Hopfield that, due to the energy and k conservation occurring in an infinite and translationally invariant medium, the Rabi oscillation would occur under resonant conditions between an exciton state and the only photon state to which it is coupled, i.e., at those exciton and photon states at the crossing point of their respective dispersion curves.

While experiments previously reported have dealt with the optical response of the coupled exciton-cavity photon mode as seen through reflectivity or transmission, it is important to evaluate how much the strong-coupling regime modifies the luminescence process, both for fundamental reasons and for applicational purposes. The situation here is quite different from atomic physics [19]: An atomic beam, having a monochromatic emission line, can only interact with a single mode of the cavity, while in a semiconductor microcavity, photons and excitons have an in-plane dispersion. During the photoluminescence process,

a number of states with different k_{\parallel} are created. It is known that this makes the analysis of bulk polariton luminescence somewhat difficult.

A sketch of a semiconductor microcavity is shown in Fig. 1. The low (high) index material is usually AIAs (GaAs), where the refractive index is $n = 2.96$ (3.54). For optical pumping it can be useful to use $\text{Al}_x\text{Ga}_{1-x}\text{As}$ ($x \approx 10\%$) instead of GaAs. The quantum wells are $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}$ and 75 Å thick, allowing experiments in reflection and transmission. 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. Figure 2 shows a photoluminescence (PL) spectrum for a series of emission angles. Figure 3 is a plot of the positions of the PL peaks as a function of the in-plane photon wave vector for three different positions on the sample. We will show that, due to the peculiarity of the emission process of the cavity polariton, the position of the PL peaks allows the direct determination of the cavity-polariton dispersion curves, as shown in Fig. 3.

In the bulk, it has been shown through a long sequence of important contributions [20–27] that excitonic polaritons have a luminescence process essentially different from the weak-coupling description, where an exciton is transformed into a photon state through a mechanism that can be modeled with a perturbative approach, such as Fermi's golden rule. In the exciton-polariton picture the emission process consists in the random-walk transfer of the polariton, regarded as a local property of the system, towards the crystal surface, and its transmission as an outside photon. Also essential is the polariton thermalization process, because both the propagation to the surface, which is characterized by the polariton group velocity and mean free path, and the transmission coefficient are very dependent on the exciton energy and wave vector. In addition, excitonic polaritons undergo a relaxation bottleneck at the resonance energy, where they can eventually connect to an outside photon state. The bottleneck leads to a peaked emission in this region [24]. Considering the contribution of generation,

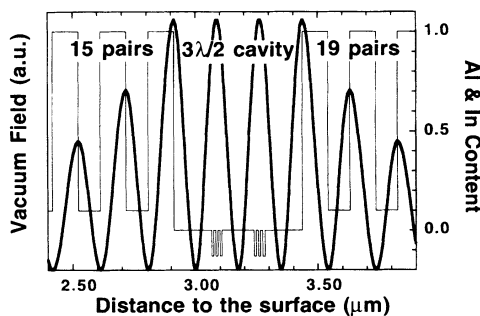


FIG. 1. Sketch of the central region of an $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{In}_y\text{Ga}_{1-y}\text{As}$ semiconductor microcavity. The thick line: vacuum field intensity. The narrow line: Al content (positive values) and In content (negative values).

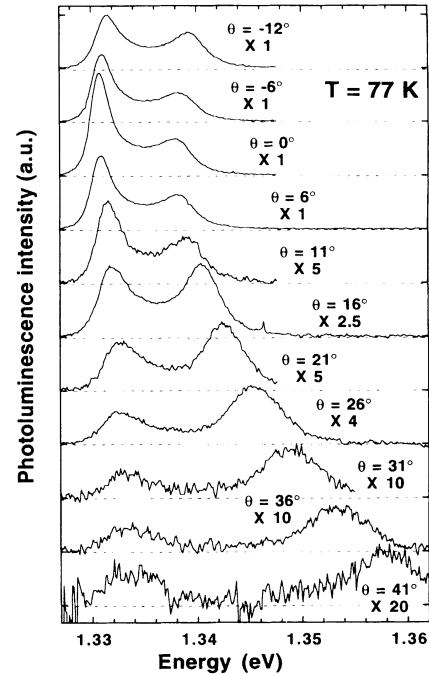


FIG. 2. Series of photoluminescence spectra at $T = 77$ K, for an emission angle from -12° to 41° . The Fabry-Pérot at normal incidence is resonant with the quantum well exciton.

energy and momentum relaxation, and recombination, it is possible, although cumbersome, to calculate an energy, angle, and position dependent polariton distribution function $f(E, \theta, z)$ [20]. It was evaluated theoretically [20] and demonstrated experimentally by resonant excitation experiments [26] that the strong-coupling regime of the exciton polariton leads to quite inefficient luminescence, even in the purest semiconductors, which can be seen as a paradox: This is a system where the transformation of a crystal excitation into a photon is as fast as it can be, but the price to pay is that this transformation is not an irreversible process anymore. Moreover, the coherent eigenstate is badly coupled to the outside photon states, because the random walk to the surface is very long compared to exciton destructive events and the transmission coefficient is most often very weak.

The bulk polariton luminescence line shape is then given by the escape rate of polaritons at the surface in the observation solid angle [27:]

$$I(E, \theta_{\text{out}}) d\Omega_{\text{out}} = \sum_{i=\text{up,lp}} f_i(E, \theta_{\text{in}}, z=0) \rho_i(E) \times v_{g\perp,i}(E) T_i(E, \theta_{\text{in}}) d\Omega_{\text{in}},$$

where the sum is over the upper and lower branch of the polariton, $f(E, \theta, z)$ is the polariton energy distribution function at the surface, ρ is the polariton density of states, $v_{g\perp}$ is the component of the polariton group velocity perpendicular to the surface, T the transmission coefficient of a polariton incident at the surface into an

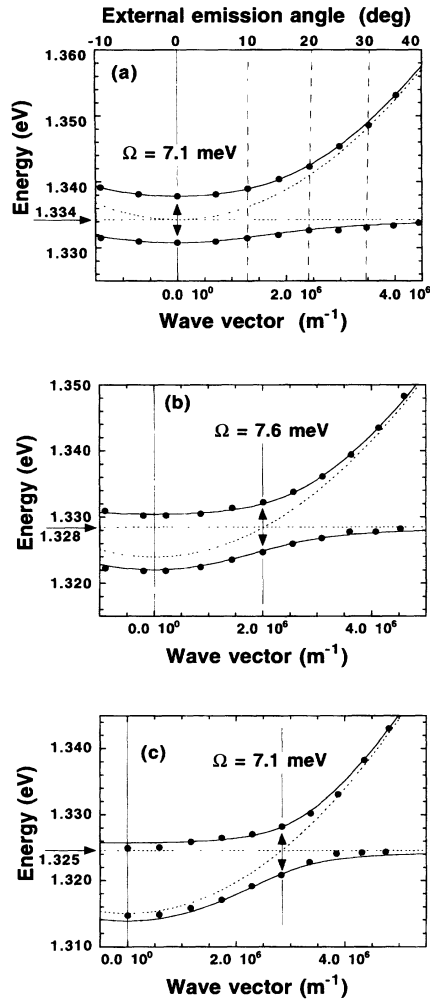


FIG. 3. Cavity-polariton dispersion curves, deduced from angle-resolved photoluminescence measurements, for different resonance conditions. (a) Resonance at $\theta = 0^\circ$ (case of Fig. 2), (b) resonance at $\theta = 29^\circ$, and (c) $\theta = 35^\circ$. The continuous lines are theoretical calculations and the dashed lines are the uncoupled exciton and cavity dispersion curves. The interaction energy Ω and exact resonance position are determined from the minimum splitting between both photoluminescence lines. An external emission angle grid is drawn on (a).

outside photon, and the propagation directions θ_{in} and θ_{out} and solid angles $d\Omega$ are related by the Snell-Descartes law. Although the energy and $k_{//}$ selection rule are already included in θ_{in} , θ_{out} , and $T(E, \theta)$, it is instructive to include them in the following form:

$$\begin{aligned}
 I(E, \theta_{out}) d\Omega_{out} &= \sum_{i=up,lp} f_i(E, \theta_{in}, z=0) \rho_i(E) \\
 &\quad \times v_{g,i}(E) T_i(E, \theta_{in}) \delta(E - E(\mathbf{k})) \\
 &\quad \times \delta(\mathbf{k}_{//} - (\mathbf{q} \cdot \mathbf{u}_{//}) \mathbf{u}_{//}) d\Omega_{in} \\
 &= \sum_{i=lp,up} I_i(E(k_{\perp} + q \sin(\theta_{out}))) d\Omega_{in},
 \end{aligned}$$

where q is the photon wave vector and $u_{//}$ a unitary vector in the direction of k . It follows that the line shape essentially depends on geometrical, static (ρ, v_g, T , etc.), and dynamic $f(E, \theta, z)$ factors. Therefore, photoluminescence of the bulk polaritons does *not* give direct information on the polariton dispersion curve without an explicit calculation of the energy distribution function.

The cavity-polariton situation is quite different: As cavity polaritons are a macroscopic property of the microcavity, the luminescence process does not involve any transport of excitation. Therefore, we can describe the luminescence process just by the knowledge of the distribution of the cavity polariton along its dispersion curve and by the outside transmission coefficient of such cavity polaritons. Because of the absence of a perpendicular wave vector the cavity-polariton case is simpler. As both the exciton and the cavity mode energy only depend on $k_{//}$, the in-plane k selection rule reduces the photoluminescence spectrum to a sum of two delta functions:

$$\begin{aligned}
 I(E, \theta_{out}) d\Omega_{out} &= \sum_{i=lp,up} f_i(E, \theta_{in}) \rho_i(E) T_i(E, \theta_{in}) \\
 &\quad \times \delta(E - E(\mathbf{k}_{//})) \\
 &\quad \times \delta(\mathbf{k}_{//} - (\mathbf{q} \cdot \mathbf{u}_{//}) \mathbf{u}_{//}) d\Omega_{in} \\
 &= \sum_{i=lp,up} I_i(E, \theta_{in}) \\
 &\quad \times \delta(E - E(q \sin(\theta_{out}))) d\Omega_{in},
 \end{aligned}$$

and therefore the emission spectrum for a fixed incidence angle should exhibit two lines whose position are *directly* related to the cavity-polariton dispersion curve and the relative intensities of which are a function of static and dynamical factors. In the presence of homogeneous or inhomogeneous broadening, the delta functions are replaced by Lorentzian or Gaussian type lines but their energy position is not changed as long as the broadening is smaller than the energy separation of both lines. This allows us to interpret Fig. 3 as dispersion curves of the cavity polariton.

Cavity-polariton dispersion curves can be calculated in different ways: (i) A classical local Lorentz oscillator model [14] can be included inside the Fabry-Pérot cavity as a dispersive medium. Although applicable to atomic physics, this model is not suitable for a semiconductor microcavity: It is independent of the Lorentz oscillator position inside the cavity, it neglects the reflections from the Lorentz oscillator layer, and it underestimates the splitting by a factor of 2 [28]. (ii) The classical Lorentz oscillator of model (i) that takes into account the reflected waves at the quantum well interfaces [15]. This is sufficient enough to solve the weaknesses mentioned for (i). (iii) An extension of model (ii) to use the transfer matrix of the quantum well layer in the nonlocal susceptibility framework [28]. (iv) Full

quantum mechanical treatment [16,17], which is usually done for a perfect lossless (i.e., closed) metallic cavity. The full quantum mechanical treatment directly gives the cavity-polariton dispersion curve vs the in-plane wave vector $k_{//}$. Models (ii) and (iii) give the dispersion curves of reflectivity, transmission, or absorption resonances in an open cavity as a function of the angle of incidence θ , with $\mathbf{k}_{//} = q \sin(\theta)$. Models (ii), (iii), and (iv) are in very good agreement.

The continuous lines in Fig. 3 are theoretical calculations of the dispersion curve using model (ii). The fitting parameters are the Rabi splitting energy Ω (which is directly related to the exciton oscillator strength f_{osc} [11]) and the resonance energy between the exciton and cavity mode. The exact resonance condition is determined when the energy separation of both lines is minimum. The dashed lines are for the uncoupled dispersion curves. In this wave vector range the exciton can be regarded as dispersionless and the cavity mode has the usual $E(k_{//}) = \sqrt{E_0^2 + c^2 \hbar^2 k_{//}^2 / n_{\text{eff}}^2}$ dependency. As can be observed, an excellent fit is obtained for $\Omega = 7.3 \pm 0.3$ meV (i.e., $f_{\text{osc}} = 4.6 \times 10^{12} \text{ cm}^{-2}$).

In conclusion, we have shown that the knowledge of the cavity-polariton dispersion relation is essential in the understanding of the photoluminescence process and that angle-resolved photoluminescence measurements allow the determination of cavity-polariton dispersion curves. An excellent agreement with theoretical calculations is found. This situation is very different from the bulk exciton polaritons and from QW polaritons not in a cavity.

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