Room-temperature cavity polaritons in a semiconductor microcavity

R. Houdré, R. P. Stanley, U. Oesterle, and M. Ilegems
Institut de Micro- et Optoélectronique, Ecole Polytechnique Fédérale de Lausanne, CH 1015, Switzerland

C. Weisbuch
Laboratoire de Physique de la Matière Condensée, Ecole Polytechnique, F 91128 Palaiseau, France
(Received 5 October 1993)

Strong exciton-photon coupling can be achieved in a semiconductor microcavity. In this regime, the Fabry-Pérot photon mode of the cavity and the exciton electronic mode are no longer the eigenstates of the system. Both states are strongly coupled and cavity polaritons occur. The splitting of the cavity polariton is also known as vacuum-field Rabi splitting in atomic physics. We observe this regime up to room temperature. This may have important implications from a device application point of view because perturbative approaches of the light-matter interaction such as Fermi's golden rule are no longer valid. The magnitude of the splitting depends on the exciton oscillator strength and provides an accurate measurement of it.

Electronic confinement in low-dimensional structures has been widely used to tailor materials for enhanced optical and electronic properties. For complete control of the emission properties, the continuum of photon states that normally exists must also be controlled. This can be achieved by confinement of photons in microcavities[1] and photonic band-gap structures.[2] On one hand, it is more difficult to confine photons than electrons because the equivalent of a potential barrier for electrons is a high-reflectivity low-absorption material, which does not exist in the visible or near IR. However, periodic structures (e.g., distributed Bragg reflectors) can overcome this limitation. On the other hand, it is much easier technologically to quantize photons in all directions than electrons, simply because the wavelength of visible and IR photons is so much greater. A three-dimensional (3D) microcavity can be more than one order of magnitude greater in all dimensions than an equivalent quantum box for electrons.

Two regimes for photon-matter interaction can occur when the number of photon modes is reduced by a microcavity, those of weak and strong coupling. In the first one, the width of the emission can be much narrower than in free space and the spontaneous emission rate can be altered,[3] either enhanced or inhibited. The change in spontaneous emission rate has led to much recent interest in microcavity structures.[4,5] The strong coupling regime corresponds to an active medium with atomiclike transitions inside a microcavity so strongly coupled to the photon modes that the position of its energy levels is altered by this coupling. In such a regime, the normal perturbative approach of Fermi's golden rule breaks down.[6] The strong coupling regime is well known in atomic physics and has recently been observed for excitons in a semiconductor microcavity at helium temperatures.[7]

In this paper we show that the strong coupling regime can be observed up to room temperature in structures where the cavity mode is resonant with the heavy-hole exciton transition. We explain our observations by analogy with atomic systems. As a consequence we can directly measure the exciton oscillator strength.

The cavity-quantum-electrodynamics (CQED) treatment of a two-level atomic system resonantly coupled to a single photon mode predicts that the eigenstates of the system are no longer the photon- and atomic-oscillator states, but two mixed symmetric and antisymmetric states. The energy separation is $\Delta_n=\hbar\Omega=g\sqrt{1+n}$ where g is a coupling factor that only depends on the dipole matrix element and the cavity volume, and n is the number of photons in the cavity.[8] For zero photons, a splitting still occurs, which can be regarded as coupling between the atomic oscillator and the vacuum field of the cavity (i.e., in the absence of a driving field). This effect was first called vacuum-field Rabi splitting by Sanchez-Mondragon, Narozhny, and Eberly[8] as it is related to the textbook case of intense-field Rabi splitting[9] which, in the present case, is induced by the zero-point field fluctuations in the cavity. If several atomic oscillators are present, it can be shown[10] that the coupling constant increases as the square root of the number N of atoms $g_{n=0}(N)=g\sqrt{N}$. If the system is prepared in a pure atomic-oscillator or photon-oscillator state, it will oscillate between these two states at the Rabi frequency Ω. In a classical description, the overall system exhibits an anticrossing behavior when both oscillators are resonant, with the two split modes corresponding to the normal modes of the system.[7] In an atomic-transition language, one considers the system as undergoing a coherent evolution with a photon being absorbed by an atom, which subsequently emits a photon with the same energy and wave vector k, the photon being reabsorbed, and so on.

The conditions for observation of Rabi splitting obtained from a linear-dispersion model or CQED are the following. The maximum splitting Ω_{max} is reached when both oscillator linewidths are matched. Ω_{max} is a function of the electric dipole matrix element of the atomic transition (d) [or the oscillator strength (f) in a classical model], the number of atomic oscillators (N), and the cavity size (V_{cavity}), i.e., the length for a planar cavity or the volume for a 3D cavity:

$$\Omega_{\max} \propto \sqrt{f/V_{\text{cavity}}} \propto d\sqrt{N/V_{\text{cavity}}} \ .$$

The Rabi frequency must be larger than the damping rate of both oscillators, i.e., $\Omega_{\max} > \gamma_{\text{cavity}}$ and $\Omega_{\max} > \gamma_{\text{atom}}$. The matching condition of both oscillator linewidths expresses, provided that the conditions for vacuum-field Rabi splitting are fulfilled, that improving one oscillator only is deterimental. In other words if the cavity damping rate is too long ("too high" Q) the system is badly coupled to the outside world and the exciton-photon particle decays before being emitted out of the cavity.

A relevant physical parameter that can be used as experimental evidence of vacuum-field Rabi splitting is absorption (i.e., the dielectric susceptibility),[10] neither reflectivity nor transmission are unambiguous parameters.[11] The existence of two split levels in the cavity plus atom system is evidenced by transitions that can occur at these energies. Existence of a structure in reflectivity does not make the distinction between absorption and a change in the reflectivity-transmission balance.

Vacuum-field Rabi splitting has been a domain of interest in atomic physics for many years. In order to observe a similar effect in solid, we need the equivalent of both atomic and photon oscillators in a monolithic semiconductor structure. Semiconductor microcavity structures have very simple 1D implementations: One uses planar Fabry-Pérot (FP) microcavities where the mirrors are multiple-quarter-wave stack distributed Bragg reflectors (DBR's). It is also well known that sharp, atomiclike excitations (excitons) do exist in semiconductors at low temperature. Due to the electron-hole interaction, the oscillator strength of the electron-hole continuum in the volume of the exciton is concentrated at the exciton energy, making it equivalent to a two-level atomic system.[12] Quantum-well (QW) excitons will have the advantages of an increased oscillator strength and an increased binding energy compared to bulk ones, and it is also possible to locate them at the exact optical-field antinode positions inside the cavity. It is interesting to recall that the equivalent of vacuum-field Rabi oscillations was proposed long ago in solids. Polaritons are just the coupled modes of the exciton (acting as a two-level system) and the vacuum field. There is no need of a cavity because of the absorption-emission k selection rule of the exciton-photon pair (i.e., the exciton can only couple to one photon mode of the same energy and wave vector). The term "vacuum-field Rabi splitting" has so far been used for semiconductor microcavities in analogy to atomic physics where this effect was first observed. From a solid-state physics point of view, where dispersion also has to be considered, the term "cavity polariton" is more appropriate.

A sketch of a semiconductor microcavity is shown in Fig. 1. The low (high) index material is usually AlAs (GaAs) where the refractive index is $n = 2.96$ (3.54). For optical pumping it can be useful to use $Al_xGa_{1-x}As$ ($x \approx 10\%$) instead of GaAs. The alloy is grown as a pseudoalloy for molecular-beam-epitaxy (MBE) technical reasons. The quantum wells are $In_{0.13}Ga_{0.87}As$ and 75 Å thick, allowing experiments in reflection and transmission. In the design of the quantum well, a compromise between the limitations induced by the mismatch strain, the increasing oscillator strength, and inhomogeneous linewidth broadening when decreasing the thickness has to be made. In optimizing the structures to achieve a large splitting, another compromise has to be made with the fact that Ω increases with f but decreases with the cavity length. In practice all the QW's cannot be positioned where the field intensity is maximum. A structure factor arises from the phase difference between the different wells and thus Ω grows more slowly than the square root of the oscillator strength. The cavity of the structures that have been investigated is $3\lambda/2$ long (i.e., two usable antinode positions) with three QW's at each antinode. Fabrication of such structures is not far from the ultimate performance of MBE systems and finesses as high as 5000 have been obtained.[13] The splitting can then be studied as a function of the detuning between the FP mode and the exciton energy by making use of the fine sample inhomogeneity across the wafer.

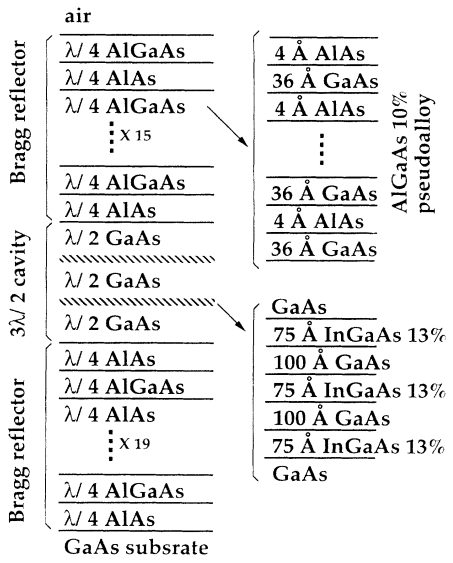

FIG. 1. Sketch of a semiconductor microcavity.

Figure 2 shows the cavity-exciton absorption spectrum, at 77 K, of the structure described in Fig. 1 when both the FP mode and the QW exciton are at resonance. The absorption (A) spectrum was deduced from reflectivity (R) and transmitivity (T) measurements. Moreover, the structure was deliberately unbalanced (i.e., the reflectivity of the back mirror is greater than the top one) so that *at resonance* $T \ll R$, then $A = 1 - R - T \approx 1 - R$. The splitting is 8.8 meV. As shown by Zhu *et al.*,[14] a linear-dispersion model can be used to describe vacuum-field Rabi splitting. In atomic physics it consists of modeling the atomic system by a set of classical Lorentz oscillators and the cavity by the standard Airy description of a FP. This model can be extended to cavity-polaritons in semiconductor microcavities. The DBR-FP is modeled by a standard transfer matrix method and a 2D exciton is included with a Lorentz oscillator dispersive dielectric con-

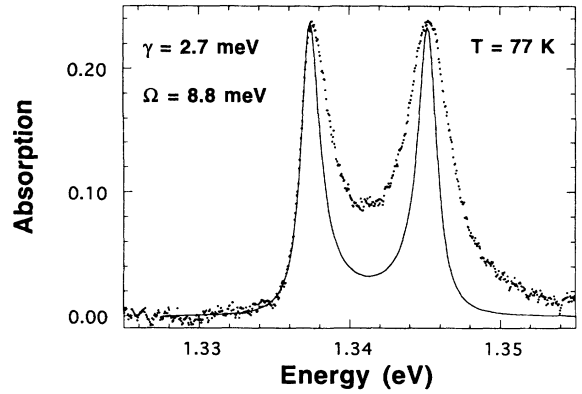

FIG. 2. 77-K absorption spectrum. The solid line is a linear-dispersion-model fit. The oscillator strength per quantum well is 4.8×10^{12} cm^{-2}.

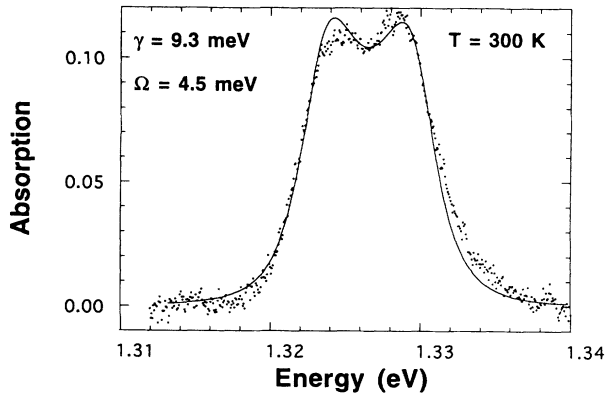

FIG. 3. 300-K absorption spectrum. The solid line is a linear-dispersion-model fit. The oscillator strength per quantum well is 4.8×10^{12} cm^{-2}.

stant:

$$\varepsilon(e)=n(e)^2=\varepsilon_\infty+\frac{fq^2\hbar^2}{m\varepsilon_0 L_z}\frac{1}{e_0^2-e^2-i\gamma e}\ ,$$

where f is the oscillator strength per unit area, q (m) the charge (mass) of the electron, L_z the QW thickness, e_0 the exciton energy, and γ the exciton linewidth. The continuous line in Fig. 2 is a fit, where the fitting parameters are the oscillator strength and the excitonic linewidth, which is assumed to be the inhomogeneous linewidth. The high-energy line is broader than the low-energy one, probably because of coupling to the continuum or excited states of the exciton. It should be noted that the observed Rabi splitting is of the order of or even bigger than the exciton binding energy. In fact, simulations including coupling to the $2s$ exciton state show a large broadening of the high-energy line. A complete fit was not attempted in this case because of too many free parameters. Therefore, the comparison to the linear-dispersion model is satisfactory for the low-energy line (especially the low-energy tail) and the amplitude of the line. A linewidth that fits the high-energy line would not have given the right amplitude and moreover would have given a low-energy tail above the experimental one. The inhomogeneous linewidth (2.7 meV) and the oscillator strength (4.8×10^{12} cm^{-2} per QW) are in good agreement with the literature.[15] It should be noted that this fitting procedure allows one of the most accurate and reliable measurements of the oscillator strength of a QW exciton. Figure 3 shows the absorption spectrum of the same nominal structure tuned so that the cavity and QW's are resonant at room temperature. The splitting is now 4.5 meV. An excellent fit is obtained with the same oscillator strength that was measured at 77 K, by adding to the 77-K inhomogeneous linewidth the homogeneous linewidth given by LO phonon scattering at 300 K ($\approx$6.5 meV).[16] No broadening due to continuum effects is observed in this case because of the smaller Rabi splitting. It should be emphasized that there are no adjustable parameters in the theoretical curve.

The observed mode splitting is not simply a filtering effect of the exciton absorption spectrum. CQED predicts that there are actually two mixed exciton-photon states coherently coupled and effects like Rabi oscillations, photon squeezing,[17,18] and dramatic change of the recombination dynamics (the usual rate equation for spontaneous and stimulated emission are no longer valid in such a system) are expected. The energy separation of these states can be larger than kT at 77 K and larger than the inhomogeneous linewidth of the same oscillator in free space. The association of quantum wells and microcavities produces an optical "hybrid" material, which exhibits a strong optical coupling, i.e., the coherent interaction of light and matter, up to room temperature where excitonic recombination does not usually occur. This might well be the "ultimate" optical material. All these microcavity concepts are just starting to influence the design of optical and electro-optic devices such as horizontal and vertical-cavity emitting lasers, electro-optic modulators, and nonlinear optic étalons. They should lead in the near future to strongly improved device performances.

This work was supported by Thomson-CSF (France), EPFL (Switzerland), and the Swiss National Priority Program for Optics.

[1]D. Kleppner, Phys. Rev. Lett. **47**, 233 (1981).

[2]E. Yablonovitch and T. J. Gmitter, Phys. Rev. Lett. **63**, 1950 (1989).

[3]P. Wittke, RCA Rev. **36**, 655 (1975); D. J. Heinzein, J. J. Childs, J. E. Thomas, and M. S. Feld, Phys. Rev. Lett. **58**, 1320 (1987), and references therein.

[4]Y. Yamamoto, S. Machida, and G. Björk, Phys. Rev. A **44**, 657 (1991); G. Björk, S. Machida, Y. Yamamoto, and K. Igeta, *ibid.* **44**, 669 (1991).

[5]*Confined Electrons and Photons*, edited by C. Weisbuch and E. Burstein (Plenum, Boston, 1994).

[6]S. Haroche, in *New Trends in Atomic Physics*, edited by G.

Grynberg and R. Stora (North-Holland, Amsterdam, 1983).
[7]C. Weisbuch, M. Nishioka, A. Ishikawa, and Y. Arakawa, Phys. Rev. Lett. **69**, 3314 (1992).
[8]J. J. Sanchez-Mondragon, N. B. Narozhny, and J. H. Eberly, Phys. Rev. Lett. **51**, 550 (1983).
[9]I. I. Rabi, Phys. Rev. **51**, 652 (1937); B. R. Mollow, *ibid.* **188**, 1969 (1969).
[10]G. S. Agarwal, Phys. Rev. Lett. **53**, 1732 (1984).
[11]R. Houdré, R. P. Stanley, U. Oesterle, M. Ilegems, and C. Weisbuch, J. Phys. (Paris) Colloq. **54**, C5-51 (1993).
[12]J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
[13]R. P. Stanley, R. Houdré, U. Oesterle, M. Gailhanou, and M. Ilegems (unpublished).
[14]Y. Zhu, D. J. Gauthier, S. E. Morin, Q. Wu, H. J. Carmichael, and T. W. Mossberg, Phys. Rev. Lett. **64**, 2499 (1990).
[15]L. Andreani and A. Pasquarello, Phys. Rev. B **42**, 8928 (1990).
[16]J. S. Weiner, D. S. Chemla, D. A. B. Miller, T. H. Wood, D. Sivco, and A. Y. Cho, Appl. Phys. Lett. **46**, 619 (1985).
[17]H. J. Carmichael, R. J. Brecha, M. G. Raizen, H. J. Kimble, and P. R. Rice, Phys. Rev. A **40**, 5516 (1989).
[18]P. Schwendimann, Z. Hradil, and A. Quattropani, J. Phys. (Paris) Colloq. **54**, C5-393 (1993).