From Fermi's Golden Rule to the Vacuum Rabi Splitting: Magnetopolaritons in a Semiconductor Optical Microcavity

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We show that magnetic quantization of the carrier free motion into discrete Landau levels for quantum wells inserted in a Fabry-Pérot microcavity allows continuous tuning of the electromagnetic interaction from the weak coupling regime (irreversible regime described by Fermi's golden rule) to the strong coupling regime (where coherent time dependence and the vacuum Rabi splitting prevail).

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This last decade has shown abundant activity in the study of atom-photon interaction in situations where only one mode of the electromagnetic field is coupled to an atomic two-level system [1]. It is now well known that in this situation the system eigenstates are two mixed electron-photon states with an energy splitting Ω depending on the atomic oscillator strength, the atom number, and the number of photons in the involved mode. As the latter is usually small, the vacuum and one-photon fields are essentially involved, and this energetic separation is usually called the vacuum Rabi splitting (VRS). It is worth recalling that, in order to obtain this "strong coupling" limit, both the electronic excitation and the electromagnetic field must have essentially discrete spectra. In contrast, if a continuum of modes of the electromagnetic field interacts with a two-level atomic system (or, conversely, if a continuum of electronic excitations interacts with a single mode), the Fermi golden rule is valid. In this case, the usual irreversible absorption or the spontaneous emission of the excited system occur, possibly modified by the actual density of electromagnetic modes: This is known as the weak coupling regime.

In solid state physics, a continuum of electronic excitations is *a priori* involved and, in spite of the large density of oscillator strength currently available, strong coupling effects should not be observed. However, in semiconductors the electron-hole interaction leads to discrete Wannier exciton bound states. A continuum of electronic states related to the exciton center-of-mass motion still exists, even in the neighborhood of the fundamental $1s$ excitonic transition, but in bulk materials the translational invariance of the electronic system restricts the interaction to only one mode of the electromagnetic field and mixed exciton-photon modes, or polaritons, occur [2]. Obviously, no such effect occurs for photon energies corresponding to the continuum of unbound electron-hole pairs.

Two-dimensional (2D) semiconductor quantum well (QW) systems are known to present enhanced excitonic features and a larger polariton splitting would be expected. Unfortunately, due to the lack of translational invariance parallel to the growth axis, the electronic system interacts with a continuum of electromagnetic modes. The very concept of polariton becomes irrelevant for wave propagation along this direction [3]. To recover the strong coupling regime and obtain an observable VRS, it is necessary, as in atomic physics, to insert the quantum well system into a microcavity which preferentially promotes an electromagnetic mode in resonance with the excitonic energy. In practice, Bragg mirrors can be grown monolithically with the QW's and form a convenient Fabry-Pérot microcavity. Splittings of a few meV have indeed been observed recently in the GaAs-GaAlAs and InGaAs-GaAs systems near the $1s$ transition of the exciton [4,5]. Note that in this case the excitonic oscillator strength per unit surface becomes the relevant quantity, in place of the atom number and atomic oscillator strength [5].

In this Letter, we address another aspect of these problems: We consider a high finesse Fabry-Pérot microcavity interacting with a 2D continuum of electronic excitations, as sketched in the left part of Fig. 1. This interaction is described by Fermi's golden rule and merely amounts to the classical absorption which damps and broadens the cavity mode. Then we add a strong axial magnetic field B which quantizes the in-plane motion of the carriers into Landau levels and discretizes their density of states, as shown in the right part of Fig. 1. If the cavity mode is between two magneto-optical transitions, we recover the high finesse "empty" cavity. If it comes into resonance with a transition, the strong coupling conditions are restored and the vacuum field Rabi splitting can be observed. We show experimentally that a magnetic field of a few teslas applied perpendicular to the layers of strained $In_{0.13}Ga_{0.87}As$-GaAs QW's embedded in a suitable Bragg-mirror cavity is sufficient to obtain a system continuously tunable from the weak coupling regime (Fermi's golden rule) to the strong coupling regime (vacuum Rabi splitting). Let us stress that these effects are essentially related to band-to-band transitions and weakly affected by

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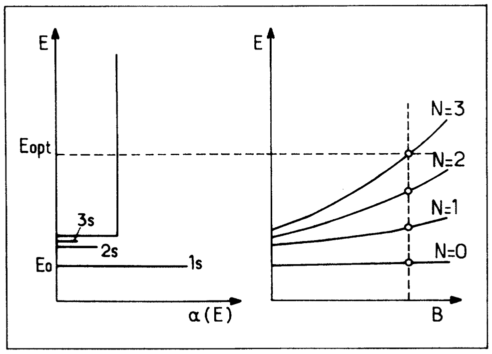

FIG. 1. Scheme of the electron-hole pair excitations in a semiconductor quantum well at zero magnetic field (left), and in presence of a quantizing magnetic field B applied along the growth axis (right). E_0 is the energy of the $1S$ Wannier exciton ground state.

their excitonic character. However, a specific excitonic effect is also obtained in these experiments, when the cavity mode is in resonance with the $1s$ exciton: In this case the diamagnetic shift of the exciton and the related increase of the transition strength allows a magnetic tuning of the VRS already observed at $B = 0$.

The sample is grown by molecular beam epitaxy on top of a GaAs substrate. It consists of a λ GaAs cavity located between two Bragg mirrors formed by stacks of $\lambda/4$ AlAs and AlGaAs layers. Three 75-Å-thick $In_{0.13}Ga_{0.87}As$ strained QW's separated by 100-Å-thick GaAs barriers are grown in the vicinity of the maximum of the electromagnetic field in this λ cavity [6]. The multiplication of QW's (three in our case) significantly increases the transition oscillator strength, and a VRS $\Omega \approx 4$ meV is observed near the excitonic absorption edge. A specific advantage of compressively strained layers in the context of these investigations is the increased splitting of the heavy and light hole levels, which produces in-plane valence band dispersions much simpler than usual: In the energy range of interest, the "heavy" hole band essentially has a parabolic in-plane dispersion with an effective mass $m_H = 0.09m_0$ comparable with the electron effective mass $m_e = 0.07m_0$ [7]. This in turn implies that the magneto-optical transitions obey a simple $\Delta N = 0$ selection rule, where N is the Landau level index, and form a simple fan chart. The fundamental gap of the QW's is near 1350 meV, which is far under the absorption edge of GaAs (1520 meV at low temperature). In order to sweep over various resonances and to follow their magnetic-field dependence, some tunability of the optical mode is required. This is obtained by an interruption of the rotation of the sample during part of the growth of the cavity layer, which gives rise essentially to a unidirectional variation of the cavity thickness across the wafer. There also exists some dispersion of the QW's composition. These gradients were measured directly by 2 K photoluminescence (excited with a He-Ne laser, far above the relevant energies) and by reflectivity measurements. We get dispersions of -3.4 meV/mm for the QW gap and $\approx +2$ meV/mm for the cavity mode energy. The various resonances and their magnetic-field dependences were obtained experimentally by recording systematically the reflectivity spectra for a 1 T field increment and a 0.5 mm increment of the spot position along the $\approx$2-cm-long sample. For clarity of discussion, we express the experimental results in terms of the detuning energy between the optical mode $E_{\rm opt}$ and the fundamental excitonic level E_0 at vanishing B at the same sample point, $\delta = E_{\rm opt} - E_0$. The reflectivity spectra were measured using an Ar^+-pumped Ti:sapphire laser in the 870–940 nm range, with the sample in the heart of a superconducting magnet. The temperature was about 2 K and B could be varied up to 9 T. According to the beam geometry, the size of the laser spot on the sample was 300 μm. In this asymmetric Bragg-mirror structure, the transmission of the system is low, even at resonance; therefore reflectivity spectra provide the same information as absorption spectra [5].

Figure 2 shows selected experimental spectra illustrating the transition between the weak and strong coupling regimes. When $\delta < 0$ [$E_{\rm opt} < E_0$, column (a)], the reflectivity peak is related to the optical mode in the nonabsorbing or empty cavity, and its observed width $\Gamma_{\rm opt} = 1$ meV. When $\delta \approx 0$ [column (b)], resonance between the ground state exciton and the cavity mode occurs and the VRS appears. The Rabi splitting Ω is 4 meV for $B = 0$ and each split line is about 3 meV broad. When B is increased, Ω increases up to 6.5 meV at $B = 7$ T. This reflects the enhancement of the exciton oscillator strength as the magnetic field progressively shrinks the bound state wave function. The asymmetry of the doublets observed in column (b) is the consequence of a finite detuning, due to the finite number of data points. Fits show that the VRS estimate is not significantly affected by this remanent asymmetry. Finally,

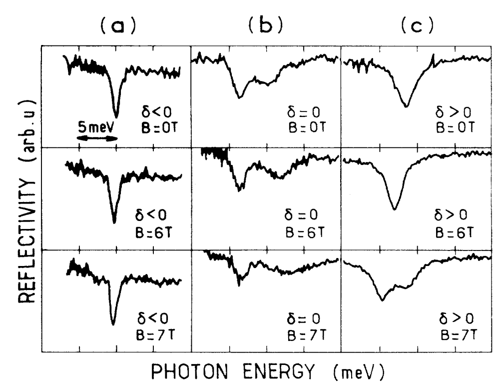

FIG. 2. Selected reflectivity spectra for various detuning energies δ and various magnetic fields B.

when δ is positive and large enough to be in the conditions of Fig. 1 [$\delta \approx 30$ meV, column (c)] only one reflectivity line is observed at $B = 0$, corresponding to the classically broadened optical mode. The increase of the resonance width with respect to the $\delta < 0$ case is due to the absorption by the continuum of unbound electron-hole pair states. The observed 3 meV width is consistent with the calculated value of the absorption probability for the three strained quantum well system [8], $\alpha \approx 1.5 \times 10^{-2}$. When B increases, the width of the resonance decreases due to the decrease of the absorption coefficient out of a magneto-optical transition (conversely, it increases due to the increase of absorption coefficient at the magneto-optical transitions) [9]. Finally, when the magnetic field is large enough (here $B > 6$ T), the magneto-optical transitions become well separated: When the cavity mode is in resonance with the discrete magneto-optical transition between the $N = 1$ conduction and heavy hole Landau levels, a new VRS is observed.

The energies of the reflectivity peaks as a function of the detuning energy are shown in Fig. 3. Whereas at low field only the VRS near the fundamental $1s$ exciton is observed, two additional VRS's are seen for $B > 6$ T, at energies corresponding to magneto-optical transitions involving the second ($N = 1$) and third ($N = 2$) Landau levels, with corresponding VRS's Ω_1 and Ω_2 of the order of 3 meV at $B = 6$ T. This is more easily seen in Fig. 4, where the energies of the reflectivity doublets at resonance are plotted as a function of the magnetic field: The fan chart of the magneto-optical transitions clearly shows up in this plot [10]. However, it should be stressed that the observed energy separations do not directly reflect the

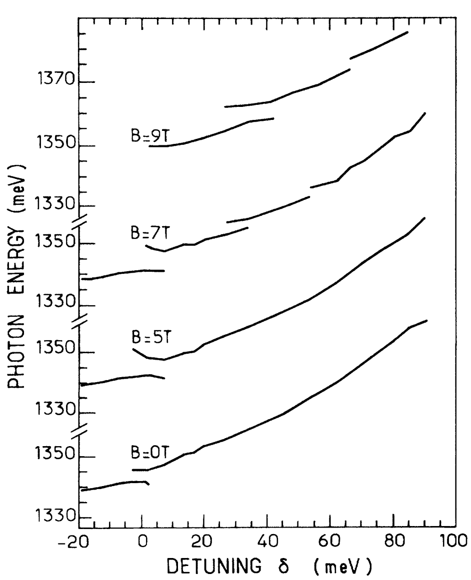

FIG. 3. Energies of the reflectivity peaks versus the detuning energy δ, for various values of the magnetic field B.

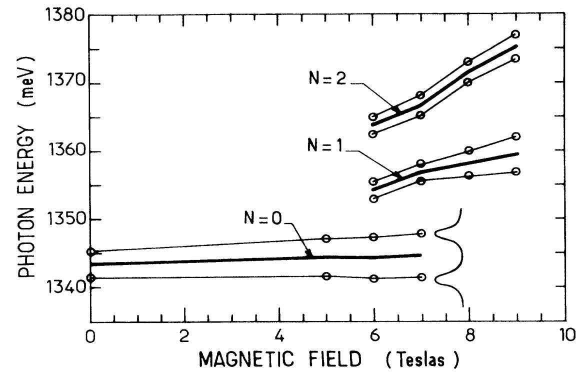

FIG. 4. Energies of the reflectivity doublets at resonance observed in Fig. 3, plotted against the magnetic field B (open circles). The solid lines are guides to the eye showing the fan chart of magneto-optical transitions.

Landau level energies, because the corresponding spectra are not measured at the same point on the sample. Taking into account the band gap gradient to correct the data of Fig. 4 yields a separation between the $N = 1$ and $N = 2$ transitions which increases by 3 meV/T. This is in good agreement with magnetophotoluminescence excitation data measured on a reference sample with equivalent quantum wells but without mirrors, and compares fairly well with the value $\hbar\omega_c/B = \hbar e(1/m_e + 1/m_H) =$ 2.9 meV/T calculated with the above-mentioned effective masses [7]. Similar to the ground state VRS Ω_0, the VRS's Ω_1 and Ω_2 associated with the transitions between the upper Landau levels increase monotonically with the magnetic field, as the Landau level degeneracy $g = 2eB/h$ (hence the transition oscillator strength) increases.

In the following, we discuss these results more quantitatively using calculated reflectivity spectra obtained in an adaptation of the semiclassical linear dispersion model of Zhu *et al.* [11], which allows a natural and convenient computation of the reflectivity spectra by a transfer matrix method [5]. In this model, the excitonic absorption line has a Lorentzian line shape with a width Γ_e and an oscillator strength f_e. This model is known to give the same results as the quantum calculation provided the probing of the system is weak. Fits of the $B = 0$ spectra yield $\Gamma_{\text{opt}} = 1$ meV and $\Gamma_e(B = 0) = 5$ meV for the $1s$ exciton. The width of each split line at resonance (3 meV) is the average of these values. When the VRS is large enough compared to the linewidths, Ω simply depends on the total oscillator strength and reaches its maximum value. When the two modes have a noticeable overlap, Ω decreases and tends to collapse [11]. In other words, for given values of linewidths, the VRS only exists above a threshold value of the oscillator strength. We find that the $\Omega_0(B = 0) = 4$ meV VRS actually corresponds to an intermediate situation, with a partial overlap of the modes. The fit yields an excitonic oscillator strength [12] per quantum well $f_e = 3.5 \times 10^{12}$ cm^{-2}, comparable with previous measurements [5]. The increase of Ω_0 with B is due to an increase of the 1s exciton oscillator strength resulting from the magnetic-field-induced shrinkage of the excitonic wave function. Assuming a constant Γ_e, the measured value $\Omega_0(B = 7\ \text{T}) = 7$ meV is nicely fitted with the value $f_e = 7 \times 10^{12}$ cm^{-2}. These values then correspond to an almost complete separation of the doublet. The factor 2 increase of the 1s exciton oscillator strength, associated with an equal increase of the exciton binding energy, is consistent with the theoretical value obtained with a simple variational description of the exciton wave function [13].

This model also provides, at least qualitatively, a criterion for the observation of VRS's Ω_1 and Ω_2. If we neglect the excitonic corrections, which are much smaller for the excited levels, the oscillator strength of the magneto-optical transitions is simply related to the zero-field interband absorption probability α by $f_e(B) = (4\pi\varepsilon_0 n m_0 c/2\pi^2\hbar e^2)\alpha\hbar\omega_c$, where n is the optical index. This simply means that the oscillator strength of a magneto-optical transition results from the squeezing of the band-to-band absorption over an energy interval $\hbar\omega_c$. Assuming that the width of the magneto-optical transitions remains of the order of 5 meV, as for the fundamental transition, the threshold $\Omega_i \geq (\Gamma_e + \Gamma_{\text{opt}})/2 = 3$ meV to observe the Ω_1 and Ω_2 splittings is $f_e > 1.5 \times 10^{12}$ cm^{-2}. This corresponds to $B > 10$ T whereas our observations indicate $B > 6$ T. The order of magnitude is satisfying, and the excitonic corrections for the upper transitions can account for at least part of the discrepancy. It is worth noticing that the observed VRS's also show a weak dependence on the Landau level index N, which is not surprising since the electron-hole excitonic binding energy and the Landau level broadening Γ depend somewhat on N.

In conclusion, we have evidenced a continuous transition between the weak and strong coupling regimes for the interaction between light and matter in a semiconductor microcavity. A magnetic tuning of the vacuum-field Rabi splitting in the latter case is also shown. In our experiments the photon mode is always discrete, and the electronic excitation is changed from a two-dimensional continuum to a series of discrete magneto-optical transitions. A "mirror" situation could be investigated in atomic physics by continuously changing the finesse of the microcavity and changing the field modes from a continuum to a single mode while obviously keeping discrete two-level excitations. Analogies and differences between these situations certainly deserve further examination, in particular with respect to the roles played by the spatial degeneracy and the in-plane translational invariance.

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