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Room temperature exciton-photon Rabi splitting in a semiconductor microcavity

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Strong exciton-photon coupling regime can be achieved in a semiconductor microcavity up to room temperature. In this regime, the Fabry-Pérot photon mode of the cavity and the exciton electronic mode are no longer eigenstates of the coupled system. Both states are strongly coupled and vacuum field Rabi splitting occurs. Analogies with atomic physics, conditions for strong coupling, experimental results and their implications are discussed.

1. Introduction.

In bulk semiconductors, radiative recombination can occur over a wide range of momentum (k) states. The dispersion of momentum with energy and the thermal distribution of carriers at room temperature results in broad emission bands. In contrast, by confining electrons and holes in low dimensional structures their energy levels become quantized and the oscillator strength is concentrated in energy. The development of low dimensional structures has progressively moved from quantum wells (QW) to quantum wires (QWW) towards quantum boxes (QB). The goal of electronic confinement is to have discrete atomic like energy levels separated in energy by a factor larger than kT and this can be achieved in QBs. However there are both *extrinsic* and *intrinsic* limitations on the optical properties of QBs. It can be shown that a bottleneck occurs in the relaxation of injected carriers in QB and this explains the observed collapse in photoluminescence of most of the quantum boxes fabricated so far [1] (although some recent experiments evidence reasonable quantum yields at low temperature, possibly at the expense of response time). Thus it seems that one might have to give up on obtaining sharp luminescence and gain curves due to exciton effects with fully quantized interband transitions. Fortunately, a new possibility is offered by the photon quantization in microcavities. The basic principle is simple: electrons and holes in energy bands can recombine at the various energies allowed by carrier population and momentum conservation ("vertical" transitions) because there exists under usual conditions a *continuum of photon states*. If the active material is placed in an optical cavity where only a single photon mode has a spectral overlap with the emission

band, radiative recombination can only take place in that photon mode. The emitted light has the spectral characteristics of the photon mode, i.e. can be extremely narrow as determined by the photon lifetime in the cavity. Like for electron states, the photon mode quantization can be achieved in 2D, 1D or 0D.

2. From vacuum field Rabi splitting in atomic physics to semiconductor microcavities.

In the field of atomic physics the influence of photon quantization due to optical cavity resonances has long been a matter of interest [2-10]. This was recently revived by the observation of the deterministic, coherent behavior of single atoms interacting with single photon modes, leading to so-called vacuum field Rabi oscillations. The area, also called the strong coupling regime, has steadily developed over the years because of the information it has provided on many fundamental aspects of quantum electrodynamics (QED) i.e. the interaction of atoms with electromagnetic fields. It is currently called cavity quantum electrodynamics (CQED), and has implications on fundamental quantum measurement theories ("Schrödinger cats") and to photon squeezed states. For the remainder of this paper we concentrate on this strong coupling regime, referring the reader to several recent publications on the weak coupling regime in semiconductors [11].

2.1. Atomic physic.

The usual multiple pass interference description of the symmetrical Fabry-Pérot (FP) resonator shows that the resonant photon mode, for which all reflected waves in the cavity are in phase, has an intensity, at the antinode position, increased by a quality factor $Q = (1+r)^2/(1-r^2) \approx 4/(1-R)$ where R (r) is the reflection coefficient for the optical (electric) field intensity ($R = r^2$). Conversely, the non resonant photon modes are suppressed by a factor $(1+r)(1-r^2)/(1+r^2) \approx (1-R)$. The cavity acts as if it concentrates the electric field in the cavity from all non resonant modes into the resonant one. It follows that the FP cavity can be regarded as a single photon mode system with a selectivity $\approx 4/(1-R)^2$ which grows as the square of the finesse F (mode width/free spectral range) $= \pi r/(1-r^2)$. In the microwave domain, a finesse up to 10^{10} can be achieved and an object located inside the cavity can only couple to a single electromagnetic mode. Provided the cavity is small enough these considerations are also valid for the vacuum field fluctuations.

The CQED treatment of a two level 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 anti symmetric states. The energy separation is $\Delta_n = \hbar\Omega = g(n+1)^{1/2}$ where g is a coupling factor that depends only on the dipole matrix element and the cavity volume, and n is the number of photons in the cavity. For zero photons, a splitting still occurs. It 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 J.J. Sanchez-Mondragon et al. [3]. If several atomic oscillators are

present it can be shown [5] that the coupling constant increases as the square root of the number N of atoms $g(N) = g_0 N^{1/2}$. If the system is prepared in the pure atomic oscillator or photon oscillator state, it will oscillate between these two states at the Rabi frequency Ω . In a classical oscillator description, the overall system exhibits an anti crossing behavior when both oscillators are resonant, with two split modes corresponding to the normal modes of the system [12]. 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 re absorbed, and so-on. This effect is related to the textbook case of intense field Rabi splitting [13] which, in this case, is induced by the zero point field fluctuations in the cavity.

The relevant physical parameters that are used as experimental evidence of vacuum field Rabi splitting are the emitted spectrum [3], absorption (i.e. the dielectric susceptibility) [5,9], time resolved oscillations [4,6] and photon statistics [8]. As will be discussed neither reflectivity nor transmission are *a priori* relevant parameters.

In order to observe vacuum field Rabi splitting, several conditions must be fulfilled [4,10]. Rabi oscillations must occur at a faster rate than atomic excited decay rate $1/\tau$ and the photon life time $t_c \approx mF/2\pi v$ in the cavity, where v [m] is the FP mode frequency [order], i.e. $\Omega t_c > 1$ and $\Omega\tau > 1$. If these conditions are satisfied the system is in strong coupling regime, if not the cavity acts as a reservoir and the excited state decays at a rate $1/t_{at} = \Omega^2 t_c$ [10]. This regime is the usual weak coupling regime where energy levels are weakly affected and perturbative approaches such as Fermi's golden rule are valid.

2.2. Semiconductor microcavities.

Vacuum field Rabi splitting in a semiconductor microcavity was first observed at 20 K [12]. Semiconductor microcavity structures have very simple 1D implementations: one uses planar Fabry-Pérot (FP) microcavities where the mirrors are multi quarter-wave stacks distributed Bragg reflectors (DBR's). 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. There are some fundamental differences between metallic mirrors FP and DBR-FP that are worth to mention. The first one comes from the fact that the electric field reflectivity coefficient is complex with a phase equal to 0 at midgap of the DBR instead of π (from the high index material, i.e. the cavity, point of view). Moreover the phase shift strongly depends on the energy. For a high index material cavity the anti node of the electric field are then at $x = i\lambda/2$ ($i \in \mathbb{N}$) instead of $(i+1/2)\lambda/2$. The second difference is that because of its distributed nature the $i\lambda/2$ long cavity layer is not the only factor that determines the effective volume of the cavity. Although the trends are the same, the relation between the finesse F and the quality factor, defined as an optical field enhancement Q or as a resolving power Q' are significantly different and intuitive guesses starting from the usual FP are often not correct.

Let us first examine the semiconductor as an atomic system: The usual optical transitions due to the creation of electron-hole pairs yield a distributed oscillator strength over the energy bands.

Although intense field Rabi oscillations have been observed in solids [14], it was pointed out [14] that electron-hole pair transitions, would not lead to vacuum field Rabi oscillations because the relaxation time of the carrier is much shorter than the expected Rabi frequency (i.e. $\Omega\tau < 1$). On the other hand, it is well known that sharp, atomic like 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, leading to an increased oscillator strength per unit volume (per unit surface for a quantum well) $f \approx f_{at}/a_B^3$ ($f \approx 8f_{at}/\pi a_B^2$), where f_{at} is an atomic oscillator strength and a_B is the 3D exciton Bohr radius and increases the Rabi frequency by a large factor. 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 exciton-photon pair (i.e. the exciton can only couple to one photon mode of the same energy and wave vector), in this case the splitting is very small (≈ 0.1 meV).

2.3. Linear dispersion modeling.

As shown by Zhu et al. [7], linear dispersion can be used to describe vacuum field Rabi splitting, although the actual interpretation of this is still controversial [7]. The DBR-FP is modeled by a standard transfer matrix method and a 2D exciton is included with a Lorentz oscillator dispersive dielectric constant:

$$\epsilon(e) = n(e)^2 = \epsilon_\infty + \frac{f q^2 \hbar^2}{m \epsilon_0 L_z} \frac{1}{e_0^2 - e^2 - i\gamma e}$$

where f is the oscillator strength per unit area, q (m) the charge (mass) of the electron and L_z the QW thickness, e_0 the exciton energy and γ the exciton linewidth. Simulations for 1λ long cavity, $f = 4.10 \cdot 10^{-12} \text{ cm}^{-2}$ and 15 AlAs/GaAs pairs, are shown in fig. 1 for weak coupling (fig 1a, $1/\gamma = 5$ meV) and strong coupling (fig 1c, $1/\gamma = 1$ meV). With only 10 mirror

Fig 1: Absorption (thick line), reflectivity and transmission (dashed line) of a Lorentz oscillator in a Fabry-Pérot distributed Bragg mirror microcavity. 1a weak coupling regime, 1b weak coupling regime with a non relevant splitting in reflectivity and transmission, 1c strong coupling regime that exhibits a vacuum field Rabi splitting.

Fig 2: Vacuum -field Rabi splitting as a function of the Lorentz oscillator strength.

important point was missed by Zhu et al. [7] and led to partially erroneous conclusions. Nevertheless it is possible to slightly unbalance the cavity so that the transmission is always much smaller than the reflection. In this case only and *at resonance* the reflectivity becomes an unambiguous parameter as it is equal to 1 - absorption.

The conditions for observation of Rabi splitting are very similar to the CQED calculation. The maximum splitting is obtained when both oscillator linewidths are matched and is only a function of the oscillator strength (fig. 2) and the cavity length. The Rabi frequency must be larger than the damping rate of both oscillators i.e. $\Omega_{\max} \propto (f \cdot \text{osc.}/\text{cavity length})^{1/2}$, $\Omega_{\max} > \delta_{\text{cav}}$ and $\Omega_{\max} > \delta_{\text{osc}}$. The matching condition of both oscillator linewidth expresses that, provided that the conditions for vacuum field Rabi splitting are fulfilled, improving one oscillator only is detrimental. 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.

3. Experimental results.

In optimizing the structures to achieve a large splitting a compromise has to be made with the fact that Ω increases with f but decreases with the cavity length. In practice all the QWs cannot be put where the field intensity is maximum. A structure factor arises from the phase difference between the different wells and thus Ω does not grow as the square root of the oscillator strength. A sketch of a semiconductor microcavity is shown in fig. 3. The material of the mirrors are AlAs and $\text{Al}_{0.1}\text{Ga}_{0.9}\text{As}$, the alloy is grown as a pseudo alloy for molecular beam epitaxy (MBE) technical reasons. The quantum

pairs and $1/\gamma = 2 \text{ meV}$ the system still remains in the weak coupling case (fig 1b). However this figure demonstrates that although a splitting can be observed in reflectivity and transmission no splitting is found in the absorption line which is the relevant physical parameter. Moreover high Q cavities are not necessary to obtain this case which can be regarded as a variation in the reflectivity/transmission balance because of phase shift induced by the exciton. This

Fig 3: Sketch of a semiconductor microcavity

Fig 4: Reflectivity spectrum of an empty cavity.

to quantify the performance of the cavity alone. The top and bottom mirrors have 20 and 27 pairs and the cavity is one wavelength long. There is a near perfect fit with the simulated spectrum in the 1.1-1.6 eV range (not shown). The full width at half maximum of the FP mode is 1.4 Å (0.2 meV) which leads to a finesse in excess of 3300, by far the highest value reported up to now. The residual broadening of the FP mode can be modeled by interface roughness and gives that the mean square thickness fluctuations is of the order of one monolayer, an intrinsic limit.

Figure 5 shows the absorption spectrum at resonance at 80 K of the structure described in fig. 3. The absorption (A) spectrum has been deduced from reflectivity (R) and transmittivity (T) measurements. Moreover it could be checked that at resonance $T \ll R$ in this unbalanced Fabry-Pérot structure, then $A = 1 - R - T \approx 1 - R$. The splitting is 8.8 meV. The continuous line is a fit, where the fitting parameters are the oscillator strength and the excitonic linewidth which is assumed to be the

wells are $\text{In}_{0.13}\text{Ga}_{0.87}\text{As}$ and 75 Å thick, allowing experiments in reflection and transmission. In the quantum well parameters the compromise is in between the limitations induced by the strain on the growth, the increasing oscillator strength and inhomogeneous linewidth when decreasing the thickness. The cavity of the structures that we have investigated is $3\lambda/2$ long (i.e. two usable anti node positions) with 3 QWs at each anti node. Fabrication of such structures are not far from the ultimate performance of MBE systems. 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.

Figure 4 shows a reflectivity spectrum of an "empty" (i.e. no QW) cavity fabricated

Fig 5: 77 K absorption spectrum. The solid line is a linear dispersion model fit.

inhomogeneous linewidth. The high energy line is much broader than the low energy one, probably because of the coupling to the continuum or excited states of the exciton. It should be noted that the observed Rabi splitting is of the order 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 fit 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 ($4.8 \cdot 10^{12} \text{ cm}^{-2}$ per QW) are in good agreement with the literature [16]. It can be noticed that this also gives one of the most accurate and reliable measurement of the oscillator strength of a QW exciton.

Figure 6 shows the absorption spectrum of the same nominal structure tuned so that the cavity and the QWs are resonant at room temperature. The splitting is now 4.6 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 ($\approx 6.5 \text{ meV}$). No broadening due to continuum effects is observed in this case because of the smaller Rabi splitting.

Fig 6: 300 K absorption spectrum. The solid line is a linear dispersion model fit.

4. Discussion

When one realizes that a linear dispersion model also predicts the so-called vacuum field Rabi splitting [7] it is reasonable to ask whether it is a classical or a quantum effect. One answer is that it is a classical effect in as much as spontaneous emission is [10]. Linear dispersion model mimics the CQED predictions and is helpful to design structures and extract phenomenological parameters but a full CQED model is needed to understand it. In fact this model only consists of two coupled harmonic oscillators and it is well known that the harmonic oscillator is one of the few problem for which quantum and classical mechanics gives the same results. Therefore it should not be surprising that a classical model coincides with the full CQED model as long as classical observables are concerned. We believe that vacuum field Rabi splitting is not only a simple filtering effect of the exciton absorption spectrum. CQED predicts that there are actually two quantum states and effects like Rabi oscillations, photon squeezing 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. Which effect will be the easiest to measure is

not clear and is an open challenge. Rabi oscillations are difficult to demonstrate not only because of the short oscillation period (500 fs) but also because of the preparation of the system into an initial pure photon or exciton state. Otherwise simple beating effect due to coherent pumping of two oscillators can be observed [17,18].

In conclusion vacuum field Rabi splitting results in two mixed exciton-photon states coherently coupled. 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 term "vacuum field Rabi splitting" has been used in analogy to atomic physics where this effect was first observed, from a solid state physics point of view it could as well be called cavity-polariton. 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 occur. This might well be the "ultimate" optical material. All these microcavity concepts are just starting to influence the design of optical and electrooptic devices such as horizontal and vertical-cavity emitting lasers, electrooptic modulators, non linear optic étalons. They should lead in near future to strongly improved devices performance.

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