

## SOURCES LASER ET SPECTROSCOPIES FEMTOSECONDES : TENDANCES ACTUELLES

### TRENDS IN FEMTOSECOND LASERS AND SPECTROSCOPY

# Femtosecond dynamics and non-linearities of exciton–photon coupling in semiconductor microstructures

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**Abstract.** We have studied the femtosecond dynamics of excitonic resonances in quantum well microcavities under strong excitation. Very strong non-linearities are observed, which bear clear resemblance to the non-linearities of an atomic two-level system. The fact that the excitonic system undergoes Rabi flopping and AC Stark splitting is clearly evidenced in a number of cases. Excitation induced dephasing shows an effect much stronger than the light dressing and prevents the observation of the Rabi flopping only when exciting in the continuum. Most of the experimental findings are well reproduced by a dynamical solution of the Maxwell–Bloch equations for an ensemble of two-level systems. This allows in particular understanding of the occurrence of strong coherent gain in microcavities. An exhaustive description of the experiments is given within the framework of semiconductor Maxwell–Bloch optical equations at the Hartree–Fock level. © 2001 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

**ultrafast dynamics / Rabi flopping / Coulomb correlation / excitons / coherence / non-linearities / pump-probe spectroscopy**

### *Dynamique femtoseconde des non-linéarités du couplage exciton–photon dans les microstructures semi-conductrices*

**Résumé.** Nous avons étudié la dynamique des résonances excitatives d'une micro cavité à puits quantique en couplage fort avec une résolution temporelle de l'ordre de 100 fs. De très fortes non-linéarités sont observées qui ont un comportement remarquablement similaire aux non-linéarités d'un système atomique à deux niveaux. Le couplage avec le champ lumineux se traduit par des oscillations de Rabi et par un éclatement dynamique de Stark, qui sont tous deux observés dans diverses configurations. Ce n'est que lorsque la microcavité est désaccordée de façon à mettre le mode de cavité en résonance avec le continuum du puits quantique que les effets collisionnels détruisent ces effets. Les résultats expérimentaux sont assez remarquablement reproduits par un modèle très simple permettant une solution dynamique des équations de Maxwell–Bloch pour un ensemble inhomogène de systèmes à deux niveaux. Ce modèle permet en outre de reproduire des observations expérimentales étonnantes comme le gain cohérent observés de manière transitoire dans les micro cavités. Une description plus quantitative des expériences est obtenue dans le cadre des équations de Maxwell–Bloch pour les semi-conducteurs au niveau Hartree–Fock. © 2001 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

Note présentée par Guy LAVAL.

*dynamique ultrarapide / oscillations de Rabi / corrélation coulombienne / excitons / cohérence / non-linéarités / spectroscopie pompe-sonde*

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## 1. Introduction

In the low-density regime, the optical response of semiconductor microstructures is dominated by the exciton, considered here as a simple quasi particle realized through the binding of one electron and one hole via Coulomb interaction (see, for example, [1]). Contributions of this quasi-particle to the optical response of quantum wells is dominant both in absorption and in emission, as well as for the non-linear response obtained for example through four wave mixing experiments. This bound state is particularly interesting as it behaves as a boson in the very low-density limit, and being a composite particle made of two fermions, shows very specific non-linearities linked to the contribution of the fermionic constituents.

In reality, excitons are a simple approximation for the first excited state of a semiconducting crystal. The non-linear interaction of light with a semiconductor is the result of a complex interplay between the different processes occurring within a huge number of interacting electrons [2,3]. In the next paper of this issue, as well as in different other publications [4–10], the complexity of the physical description of the excitonic related non-linearities, as well as of the experiment used to assess them has been described in quite some detail [2,11–14].

It has for example been emphasized that a proper description of excitonic optical non-linearities, intrinsically linked with the infinite range Coulomb interaction between the charge carriers, involves a hierarchy of operators up to any order, becoming more and more complex as the precision of our measurement and theoretical techniques evolves [15]. Proper truncating schemes [16,17] have to be used to describe a wide range of non-linear experiments [6,18,19]. This approach is essential when excitons and free carriers are simultaneously excited, as is usually the case when a short optical pulse is used.

Here, in order to understand the non-linearities of the system, we use an approach that is specifically suited to the case of resonant excitation at the energy of the exciton ground state. The aim of this approach is not to simplify arbitrarily the theoretical description of a complex system, but to provide an explanation allowing understanding of, in a direct and comprehensive fashion, the principal mechanisms responsible for the observed non-linearities.

For example, the different contributions to the non-linearities in quantum wells are usually traced back to the combined effects of phase space filling, screening and excitation induced dephasing [20–22] as well as higher order correlations [16,17]. These non-linearities weaken the excitonic resonances, and limit their possible use. The precise balance between the existing non-linearities and the permanence of the excitonic bound states is a topic of some importance, knowing the possible use of excitons in a number of real devices such as SEEDs [23] or micro-OPOs [24].

With a simplified quantum optics image in mind, the comparison of the exciton states with atomic two-level systems is very appealing as excitons only interact with light over a very narrow range of energies. One may wonder whether the excitons do behave approximately as two-level atoms, as far as their non-linear properties are concerned, or if they show very different non-linearities. The first observations in this field are due to Fröhlich [25], Von Lehmen [26] and Myzyrowicz [27], who evidenced the AC Stark coupling of excitons under excitation by a strong non-resonant optical pulse in the perturbative regime. We will pursue this direction of research here, and look for the similarities and differences between excitons and two-level atomic systems. Our results will be compared to a theoretical approach, based on semiconductor Maxwell–Bloch equations for the description of the excited electrons and holes and their coupling with the light field [28].

As already mentioned, Coulomb correlations between excitons, and excitation induced dephasing are expected to wash out very rapidly the excitonic resonances, before the expected non-linear effects in the

non-perturbative regime can be evidenced. However, as would be expected for a strongly excited two-level system, we have been able to observe directly different effects such as Rabi flopping [29], AC Stark splitting [30,31] or even hyper-Raman gain [32]. These observations are only possible through the use of purposely designed structures, allowing the increasing of the strength of the optical field and of the exciton light coupling and the limiting of the possible sources of dephasing by keeping a spectrally narrow enough pulse width. In such a way, the number of free electron-hole pairs excited by the light pulse is kept to a minimum.

## 2. Experimental details

The experiments we will describe here have been usually carried out in the pump and probe geometry (with the probe either in reflection or in transmission), the two beams crossing on the sample surface at near normal incidence. It is essential to properly image the different spots at the surface of the sample, and to filter spatially the probe beam (or the pump beam) so as to only report the results pertaining to a homogeneously excited region of the sample. The probe intensity is always very weak compared to that of the pump, so as to probe in the linear regime. Unless otherwise specified, the pulses are transform limited, 100 fs long, and produced directly by a titanium sapphire oscillator. The experiments are carried out in all cases at liquid helium temperature, the signal being detected by a cooled CCD camera or a PM tube behind a spectrometer.

The samples are InGaAs quantum wells with a low Indium content, and the Bragg mirrors are made of 10% AlGaAs, AlAs quarter-wave stacks grown by molecular beam epitaxy. What is very important here is the very high quality of the samples that can be produced now, limiting the importance of inhomogeneous broadening as much as possible. We used different microcavities, in the strong coupling regime, showing different qualities. The results presented here seem to be related to intrinsic effects as they are all the more easily observed in the highest quality cavities. Such cavities show, for a single quantum well, a linewidth of 100  $\mu\text{eV}$  for the two-polariton modes and a normal mode splitting of 3.5 meV [33].

We define the intensity in our experiments by reference to the saturation density computed by Schmitt-Rink et al. [20]. They estimate an exciton density  $n_{\text{psf}}$  of  $5 \cdot 10^{10} \text{ cm}^{-2}$  for saturation. The corresponding pulse saturation intensity  $I_s$  of  $2 \cdot 10^{12} \text{ photons}/(\text{pulse} \cdot \text{cm}^2)$ , is computed by taking into account the absorption of the cavity. We will use this density as a reference throughout the paper.

## 3. Rabi flopping and AC stark splitting

Since the seminal work of Lehmen [26] and Myzyrowicz [27], the question of a possible direct observation of Rabi flopping in semiconductor systems has been a long-standing debate. The question raised by some theorists being, since then, that although closely resembling to a two level system, excitonic states in semiconductors suffer from their fermionic constituents. They are responsible for both phase space filling (PSF), useful for non-linearities, and excitation induced dephasing (EID), which destroys the coherence of the system with growing density. In fact, spectrally broad short pulses are needed for the observation of strong enough effects, which means that excitons and free carriers will be created at the same time.

However, some signatures of Rabi flopping have been first reported by Cundiff et al. in [34] for resonant excitation. They measured the time resolved properties of a pulse reflected at the surface of a quantum well sample under high excitation conditions. They interpreted their results as due to Rabi flopping through detailed modeling, although no direct observation of the oscillations of the population of excitons could be given. Also, the spectral oscillations, that should associated with the temporal flopping, could not be resolved, because a spectrally narrow pulse was used for excitation.

Although some experiments on microcavities do evidence effects due to the non-perturbative regime, there has been no report for the observation of Rabi oscillations or Stark splitting [35,36] before our experiments. This, despite the fact that the observation of non-perturbative effects should be greatly improved by using microcavities. Indeed, in such a structure the strength of the optical field in the cavity

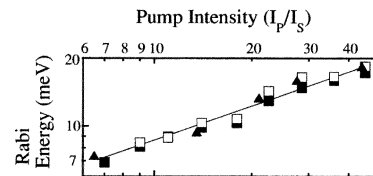
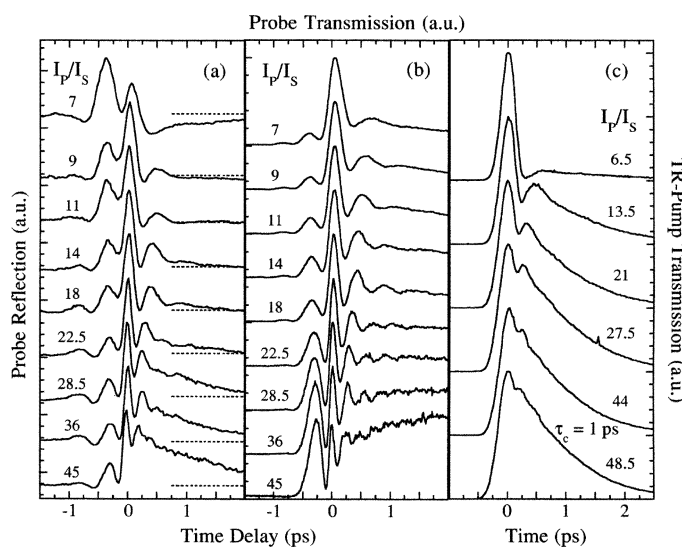
is enhanced, and at the same time, the cavity acts as a filter for the short laser pulse, which means that free carriers will not be excited. It is then possible to observe effects linked to the sole exciton non-linearities directly using a wide 100 fs probe pulse. Let us note that as the cavity filters the pulse, its time decay will get much longer (given by the cavity photon lifetime), but the rising edge will keep its ultrashort time duration. MCs containing absorbing QWs are thus particularly well suited to investigate the coherent properties of the light–semiconductor interactions in the excitonic regime.

The linear optical properties of microcavities [37–39] as well as their non-linear properties [40,41] have been studied in detail in the low-density regime using four-wave mixing and pump-and-probe experiments. It is only quite recently that the effects of strong excitation have been addressed by different techniques, giving rise to a very rich phenomenology [24,29,42–44]. We report below experiments under very strong optical pumping able to produce exciton densities well above the saturation density (these experiments have been partly described in [29–32]).

When resonantly exciting a microcavity with a strong laser pulse, we observe clear signatures of the Rabi flopping of excitons in the cavity. Such oscillations are plotted in *figure 1* in different configurations. In panels (a) and (b), the reflected and transmitted probe intensities are recorded by setting the detection energy at the position of the bare cavity mode (the experiments are performed close to zero detuning and at normal incidence).

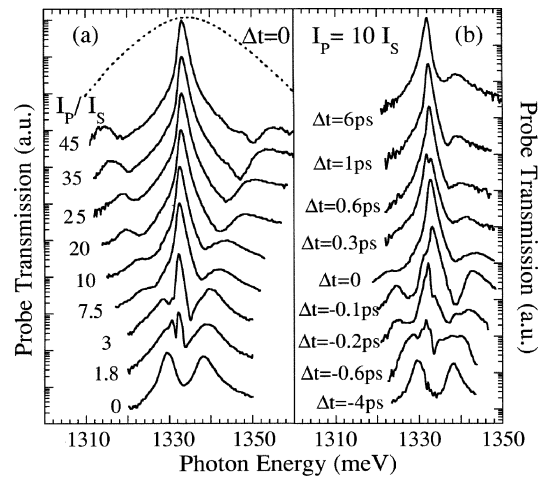
Of course, the observed effects arise for densities which are of the order of the saturation density of the exciton–cavity coupling and above, which means that the cavity goes under weak coupling regime. It is very clearly observed that the period of the oscillations gets shorter as the pulse intensity gets stronger, and that dephasing also gets shorter at the highest densities, not preventing however the clear observation of the Rabi flopping. The inverse of the oscillation period shows a simple linear dependence with the square root of the laser power (see *figure 1d*) as is expected for Rabi flopping in the simplest version [45].

To demonstrate further that these oscillations correspond to population beating, we plot in *figure 1c* the time resolved pump intensity, transmitted through the cavity (obtained by gating the pump pulse via upconversion in a BBO non-linear crystal). At high intensities, the pump is mainly reshaped by the cavity, showing a leading edge given by the pulse duration, and a trailing edge by the photon lifetime in the cavity. However, oscillations are observed in all transients, whose periods are also plotted in *figure 1d*, and fit perfectly with the period observed for the probe pulses.



**Figure 1.** Rabi flopping in a semiconductor microcavity evidenced through the variations versus pump-probe delay: (a) of the intensity of the probe pulses reflected from the surface of the microcavity; (b) of the probe pulses transmitted through the pumped MC; (c) the time resolved transmitted pump pulse. The oscillations are shown for different excitation densities in terms of the saturation intensity  $I_s$ ; (d) log–log plot of the Rabi energy associated with the different beating periods in (a)–(c), as a function of the pump intensity. The continuous line corresponds to the square root law.

**Figure 2.** (a) Probe transmission spectra of the MC, measured using the probe pulses at zero pump-probe temporal delay, for different pump intensities in terms of  $I_s$ . The measurements have been performed at resonance between laser photons, X and the cavity mode. (b) Variations of the probe spectra as a function of the delay between pump and probe for an excitation intensity of  $10I_s$ .



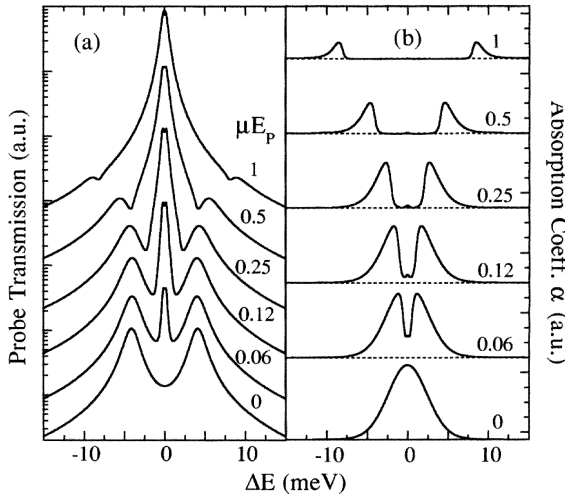
It is revealing to observe directly the spectra associated with the Rabi flopping in the microcavity. Such spectra are shown in *figure 2* for pump and probe pulses arriving at the same time on the sample ( $\Delta t = 0$ ), and for measurements in transmission (these measurements are allowed thanks to the fact that the quantum wells are made of InGaAs). With increasing pump intensity  $I_p$ , the transmission spectra evolve from the low-density polariton doublet to a practically symmetrical triplet structure (Mollow like) [46], consisting of a very strong central line and two weak lateral ones (see *figure 2*) [47].

The main peak, appearing at the center of the spectrum, corresponds to the cavity mode due to the transition of the cavity towards weak coupling regime, because of the saturation of the excitonic absorption. Strong oscillations are observed in the intermediate density domain for which we do not have a complete quantitative explanation. At high excitation intensities, the spectral separation of the lateral lines is proportional to  $I_p^{1/2}$ . This spectral separation of the lines agrees perfectly with the inverse of the oscillation period evidenced in *figure 1* (see the solid squares in the inset of *figure 1*). The spectral oscillations that we observe cannot be confused with the well-known spectral artifact (see, for example, [48]). Spectral oscillations due to this spectral artifact show a period, which depends on the time delay between the pump and the probe pulse (they are simply the Fourier transform counterpart of the two pulses exciting the sample). On the contrary, the spectral oscillations appearing in the frequency spectrum of the microcavity (*figure 2*) show an energy separation, which depends on the intensity of the pulses for the same delay.

The magnitude of the Rabi energy, extracted both from the Rabi flopping in the time domain, and from the AC Stark splitting in the frequency domain is very large. From the intracavity peak intensity  $E_p^{\text{peak}}$  for the highest pump intensity, we find a Rabi energy  $E_{\text{Rabi}} = \mu_{cv} E_p^{\text{peak}} \approx 15$  meV, which is perfectly consistent with our observations.

A very simple model can be developed, in order to reproduce the spectral oscillations. It consists of the steady-state solution of Bloch equations for a two-level absorber. This allows us to calculate approximately the susceptibility of the inhomogeneously broadened X transition, provided that only the  $1s$  exciton state is considered and that many body effects are reduced to a saturation of the oscillator strength. When such an ensemble is excited by a very strong pump pulse, a hole is formed in the middle of the distribution, and two side bands develop on either side of the unperturbed resonance. When placed into a microcavity, such a system should show an optical response simply given by the transformation of the computed susceptibility by the transfer matrix method. This is shown in the right panel of *figure 3*. It is clear that, although the details are not well reproduced, the main features of the observed spectra correspond quite well to such a simple image.

In reference [28], we solve the Hartree–Fock semiconductor Bloch equations, projecting into the  $1s$ -exciton state. In order to go further, Quochi et al. [29] have developed a dynamical solution of the



**Figure 3.** (a) Calculated probe transmission spectra for different pump intensities indicated in units relative to the highest density. (b) Absorption coefficient of a broadened two level system versus probe to resonance detuning.

semiconductor Bloch equations for an ensemble of interacting two level systems. The second order Maxwell's equations are here replaced by an effective first-order oscillator equation for the internal field mode. They also use the quasi-mode approximation for the description of the relation between the intracavity field and the external field [49]. In such a case, the susceptibility of the system is given by the resolution of three coupled differential equations describing the evolution of respectively the carrier density, the polarization and the optical field:

$$\begin{aligned}\dot{n}_X(t) &= \frac{iV}{\hbar} \{ \Omega_c(t) p_X^*(t) - \Omega_c^*(t) p_X(t) \} \frac{n_X(t)}{T_1} \\ \dot{p}_X(t) &= \frac{i}{\hbar} (E_X - \hbar\omega_p) p_X(t) + \frac{iV}{\hbar} \Omega_c(t) \{ 1 - 2n_X(t) \} - \frac{p_X(t)}{T_2} \\ \dot{\Omega}_c(t) &= \frac{i}{\hbar} (E_X - \hbar\omega_p) \Omega_c(t) + \frac{iV}{\hbar} \int dE_X G(E_X, E_{X0}) p_X(t) + \frac{\Omega_c(t)}{2\tau_c} + \tilde{g} \Omega_{\text{ext}}(t)\end{aligned}$$

Where  $p$  is the polarization,  $n$  the population and  $\Omega$  the Rabi energy (all dimensionless). The Rabi energies  $\Omega_c(t)$  and  $\Omega_{\text{ext}}(t)$  are related either to the intracavity field  $\varepsilon_c(t)$ , or to the external field  $\varepsilon_{\text{ext}}(t)$  (coupled by the coupling rate  $\tilde{g}$ ) through the relations:

$$\Omega_j(t) = \frac{\mu \varepsilon_j(t)}{V}, \quad j = c, \text{ext}$$

$V$  being the coupling energy between the two-level systems and the cavity mode [12]. The energy of the excitons is  $E_X = \hbar\omega_X$  and that of the cavity photons  $E_c = \hbar\omega_c$ , the lifetime of the photon in the cavity being  $\tau_c$ . The distribution of excitonic energies is given by a Gaussian function  $G(E_X, E_{X0})$ .  $T_1$  and  $T_2$  are the longitudinal and transversal relaxation times of the system. The many-body effects are implemented phenomenologically on the basis of the theory developed by Schmitt-Rink et al. [20]. At first order, the increase of the exciton density decreases the oscillator strength and blue shifts the exciton resonance. This approximation is equivalent to the local field approximation used by Wegener et al. [54]. This is introduced in terms of the dimensionless exciton density  $n_X$ , and dimensionless saturation density  $n_{\text{PSF}}$  through:

$$\mu(n_X) = \mu_0(1 - n_X) \quad \text{and} \quad \delta E_X(n_X) = 2n_X E_B$$

We also account for the excitation induced dephasing in the simplest possible form by writing:

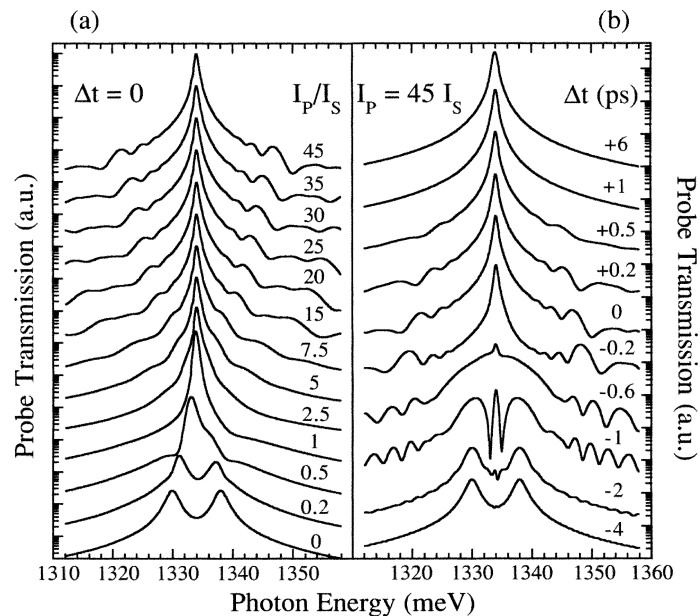
$$\gamma(n_X) = \gamma_0 + 2n_X(\gamma_{\text{PSF}} - \gamma_0) = 2\hbar/T_2(n_X)$$

From these equations, the linear susceptibility of the ensemble with respect to the probe beam is obtained through the Fourier transforms of the changes in the polarization over the changes in the cavity field:

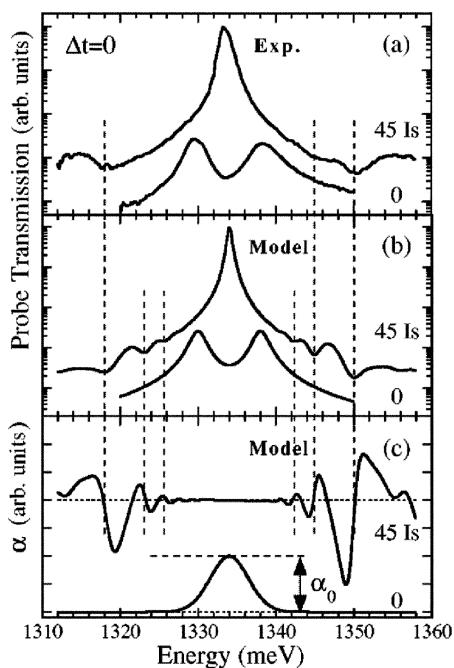
$$\chi(\omega, t) = \frac{\delta\tilde{P}(t, \Delta t)}{\delta\tilde{\Omega}_c(t, \Delta t)}$$

This susceptibility is then transformed through transfer matrix calculation at normal incidence to account for the effects of the cavity. The transmission or reflection spectra are plotted in *figure 4* in two interesting configurations. In *figure 4a*, the probe transmission at zero time delay is plotted, as a function of the pump intensity expressed in units of the saturation intensity (outside the cavity). The first phenomenon that is evidenced is a collapse of the low-density polariton splitting of the cavity, due to the saturation of the excitonic resonance. This goes with a large increase of the transmitted intensity. The slight asymmetry of the collapse is due to a subtle interplay between the loss of oscillator strength and the blue shift of the exciton resonance. Basically, the cavity goes under weak coupling for  $0.5I_s$ , but at this density already, some indication of the AC Stark bands appear. These sidebands develop and get more separated, on either side of the cavity resonance, as the intensity of the pump pulse is further increased. A series of sidebands is observed due to the slow decrease of the intensity of the pump in the cavity, following the photon lifetime.

In *figure 4b* are presented the results of the dynamical calculation for the highest pump intensity, as a function of the delay between the pump and the probe. Already at negative delays, some oscillations are clearly visible, which correspond to the interference between the polarization left by the probe in the cavity, and the pump. As discussed above, these oscillations, the so-called spectral artifact, are well documented and their period is simply the inverse of the time delay between pump and probe. Their intensity is strongly increasing towards zero delay as the probe polarization in the cavity only has a lifetime of 1 ps. Such oscillations can be resolved in some of our experimental spectra (not shown here), but are in general not observable.



**Figure 4.** Results of the dynamical solution of Maxwell–Bloch equations. (a) Calculated probe transmission spectra at zero delay as a function of the pump intensity. (b) Probe transmission as a function of delay for the highest pump intensity.



**Figure 5.** Comparison between the experiments and the dynamical solution of the Maxwell–Bloch equations for an ensemble of two level systems in a microcavity. (a) The experimental spectra at zero density and at very high excitation ( $45I_s$ ,  $I_s = 2 \cdot 10^{12}$  photons/(pulse·cm<sup>2</sup>)). (b) The computed response for the same conditions. (c) The susceptibility of the quantum well, before application of the transfer matrix, showing very strong oscillations. The vertical lines mark the spectral positions of the Stark sidebands; the horizontal line marks zero absorption.

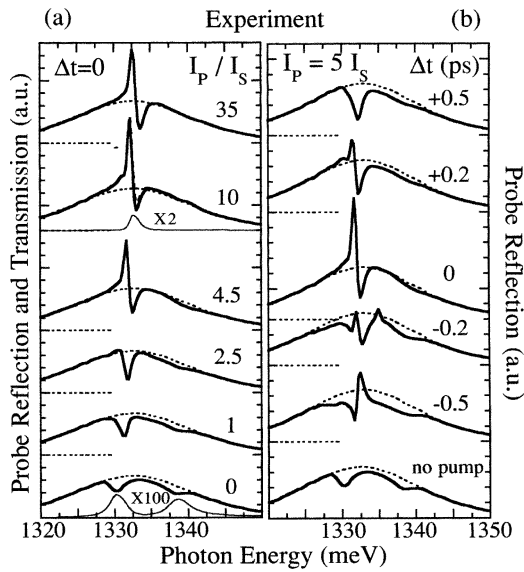
The cavity polaritons already collapse for a delay of about  $-0.6$  ps due to the very large pump intensity, and the AC Stark bands clearly appear for a delay of  $-0.2$  ps. Their energy position is not constant due to the decaying intensity of the pump in the cavity. Within the first picosecond, the sidebands completely disappear, leaving only the strong cavity mode due to the large population of incoherent excitons in the cavity.

The calculated spectra, despite the simplifying assumptions made for the computation, agree quantitatively very well with the experimental ones. A further comparison is performed in *figure 5* where we detail the changes between the lowest and the highest excitations for the experimental and calculated spectra (in *figures 5a* and *5b*), as well as the corresponding susceptibility in *figure 5c*. The agreement, once again is very good. The observed Stark bands do correspond to the calculated ones, although the fine oscillations predicted by the theory are not fully resolved in the experiment.

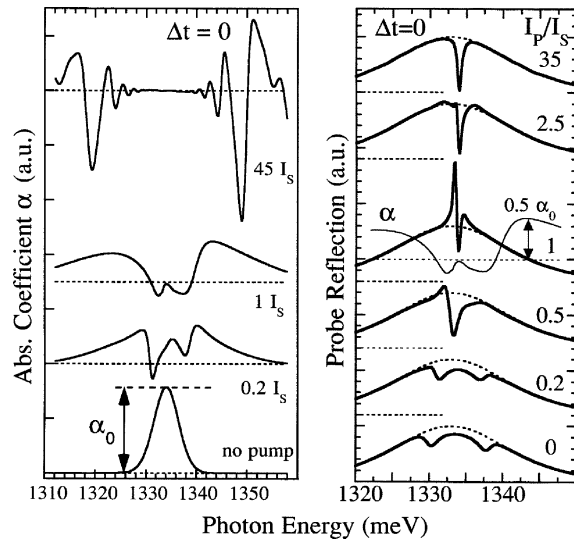
It is worthwhile noting that we observe Rabi flopping for times of the order of one ps, or more, although the expected  $T_2$  dephasing time due to excitation induced dephasing should be much shorter. In fact, we can experimentally get a very good estimate of this dephasing time from the observed linewidth, which is 5 meV at the highest densities. This gives a dephasing time at high density of 0.25 ps, which we have introduced in our model. The model then predicts in such a case that oscillations will be observed for 1 ps, because of the long duration of the pulse in the microcavity. In the experiments of Norris et al. [50], the cavity lifetime is too short (0.25 ps) compared to the dephasing time, to allow the observation of the Rabi oscillations.

#### 4. Coherent gain

The above description, which allows interpretation of the observed spectra, is also able to provide a reasonable explanation for the coherent gain observed in the same excitation conditions. This gain is observed by looking at the reflected part of the probe beam rather than the transmitted part [32]. In this experiment, whose results are depicted in *figure 6*, we observe the modifications of the reflection by the cavity polaritons under strong excitation, for zero pump and probe delay, as a function of the pump intensity (a). In *figure 6b*, we plot the reflected probe beam as a function of delay with the pump for an excitation intensity of  $5I_s$ . At the lowest intensities, we observe the expected polariton doublet. At high



**Figure 6.** Reflection spectra of the weak probe pulses for different pump-probe delays  $\Delta t$ : (a) measured for  $I_p = 5I_s$ ; (b) calculated for  $I_p = I_s$ . The smooth dashed curves represent the spectra of the incident probe pulses, while the horizontal dashed lines mark zero reflection.



**Figure 7.** Comparison between the computed quantum well susceptibility (left panel) and the corresponding reflectivity response of the microcavity. The strong oscillations that appear in the susceptibility only appear close to  $I_s$  because of the filtering effect of the cavity.

densities and long delays, as expected, the cavity goes under weak coupling, and only the cavity mode is observed. However, at zero time delay, a strong gain is observed close to the cavity energy (the AC Stark sidebands cannot be resolved on a linear scale).

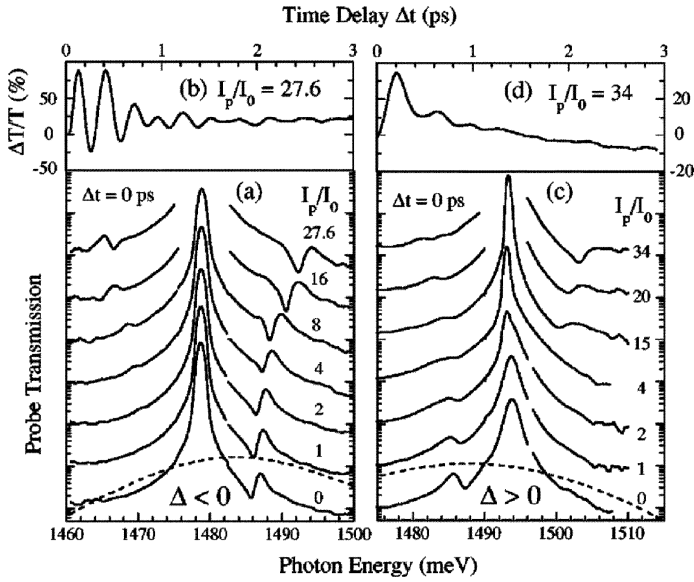
This gain only occurs during the coherence of the pump pulse, and can be interpreted as hyper-Raman gain [47]. It can be as large as a factor of three for a single quantum well, i.e. much larger than observed without the microcavity [51].

In order to understand more precisely the origin of this gain, it is very instructive first to compare the experiment to computed spectra (corresponding to the experiment of *figure 6a*). The left panel of *figure 7* shows the computed susceptibility of the quantum well for different excitation densities. The right panel of the same figure shows the corresponding spectra after the filtering effect of the microcavity. These spectra reproduce quite well the observed effects; with the difference that the gain observed at very high densities is not reproduced by the calculations.

The origin of the gain peak is more easily understood when looking at the calculated susceptibility of the quantum well. At low intensities, the pump penetrates the cavity at the polariton energies. A strong pump ( $0.2I_s$ ) creates two dips in the absorption spectrum, which are very slightly asymmetric due to the combined effect of saturation and blue shift on the cavity resonance. At higher intensity ( $I_s$ ), Stark sidebands are already present and gain shows up only within the pass band of the cavity. As the cavity mode builds on the low energy side, it will correspond to the lower dip, showing therefore a gain amplified by the effect of the cavity. At very high intensity ( $45I_s$ ), the gain appears only far from the cavity mode and only gives rise to small undulations.

## 5. Rabi flopping for strongly detuned cavities

What all these experiments evidence is that, when only exciton transitions are excited, the semiconductor behaves approximately as an ensemble of two level systems broadened by disorder in the quantum wells.



**Figure 8.** AC Stark splitting and Rabi oscillations observed, in the case of very strong detuning (both positive and negative detuning). The right part of the figure shows that this is also the case when the excitation energy is resonant with the continuum, although with faster dephasing.

The Coulomb interactions do not fully destroy such effects as excitation induced dephasing is obviously longer than the period of the Rabi flopping that can be achieved. We also know that exciting free carrier pairs, together with excitons, will tend to increase dramatically the dephasing rate. Once again, a microcavity is a system of choice to study such effects, because the cavity mode can be detuned at will with respect to the excitonic resonance. Therefore it is possible to excite free carriers with a well-defined excess energy. We report the results of a pump probe experiment for a strongly detuned microcavity in *figure 8* [30]. Please note that the sample used for this part of the study is of much higher quality than the previous one, the linewidth at low density being close to 100  $\mu\text{eV}$ .

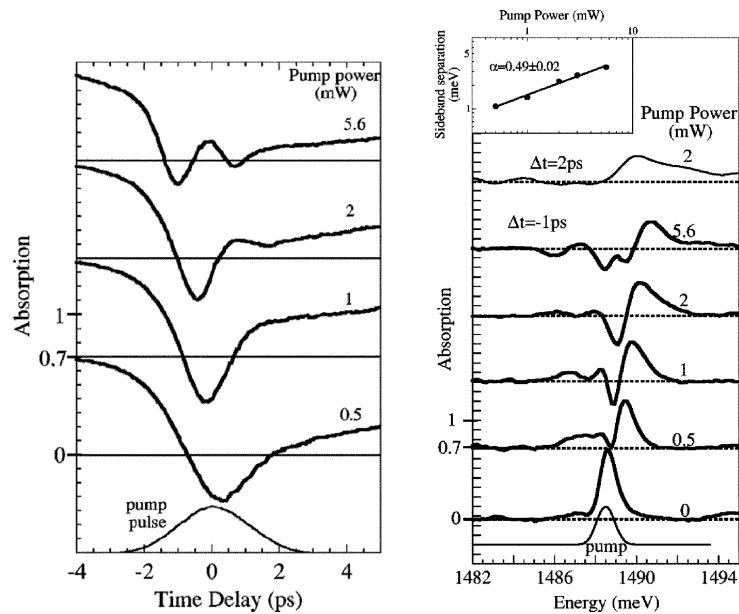
In the left part of the figure, we report the  $\Delta t = 0$  spectra as well as the temporal oscillations at the cavity energy for the case of strong negative detuning. Strong Rabi flopping as well as AC Stark bands is evidenced in the two panels. This indicates that, although the pump is now strongly detuned from the exciton energy, the optical field is enhanced enough in the cavity to be able to bring the system to the non-perturbative regime. The right panels show the same effect, when the cavity is now strongly positively detuned. In the case of *figure 8*, we made sure that the cavity energy is slightly above the continuum energy. This is clearly shown in the spectra through the increase of the upper polariton linewidth. Nevertheless, Rabi flopping and AC Stark bands are also observed in such a case. However, it is clear that the dephasing rate is much larger now as the Rabi oscillations are very rapidly damped. If we detune the cavity further, the oscillations rapidly disappear.

## 6. Rabi flopping in a single quantum well without cavity effects

In microcavities, the non-linear excitonic effects are more easily observed because of a double effect: the increase of the strength of the optical field due to the confinement of the electromagnetic field in the cavity, and the filtering effect of the same cavity. Nevertheless, it should be possible to obtain the same effects in the case of bare quantum wells, provided a strong enough pulse is sent on the sample, and direct excitation of free carriers is avoided. Preliminary experiments have been reported by Schultzgen et al. [52], who observed oscillations by probing at the light-hole-exciton energy.

We have specifically designed a sample that should allow the observation of Rabi flopping for a single quantum well [31]. It is indeed important to use a single quantum well as the effects directly depend on the excitation density, which will of course vary from well to well in a multiquantum well system. We have

**Figure 9.** AC Stark splitting (left panel) and Rabi oscillations (right panel) for a single quantum well.



placed the single InGaAs quantum well at the middle of a lambda thick layer placed on a Bragg mirror, made of 30 repeats of  $\lambda/4$  GaAs–AlAs stacks. Such a structure has the advantage of giving rise to a maximum of the electromagnetic field exactly at the position of the quantum well. In such a case, the absorption of the quantum well is strongly modified and amounts to 70% at the exciton energy, i.e. approximately 30 times the absorption of a bare quantum well. This absorption is not measured in the transmission geometry, but can be directly assessed in reflection thanks to the Bragg mirror, which approximately reflects 100% of the incoming light. The main differences in the experiments compared to the case of the microcavity sample are:

- due to the absence of the filtering effect of the microcavity, the pump pulse has to be tailored to match the excitonic resonance. We have used a pump pulse shaped to be 2 ps long (1 meV wide) at the energy of the exciton (1.485 eV);
- as the electric field of the optical mode is not enhanced in a way similar to a high  $Q$  cavity, the pulse energy has to be much larger, and our experiments have been limited at high density by the power available from our high repetition rate Ti:saphir laser.

The experiments indeed provide the evidence that the light dressing can be stronger than the excitation induced dephasing effects, provided free electron hole pairs are not excited. This can be first shown directly by the very strong Rabi oscillations observed at the energy of the exciton resonance. The signal goes from 70% absorption to as much as 35% gain over the onset of the pulse. Strong oscillations occur over a time period of about 2 ps (see *figure 9*). The period of the oscillations gets shorter as the power is increased, following the expected square root law as a function of the incident power. Simultaneously, the excitonic spectrum is strongly modified. It is shown in the right panel of *figure 2* for a delay of  $-1$  ps between the pump and the probe. This delay has been chosen because it corresponds to the maximum of the effects observed in the time domain. The susceptibility goes through a series of minima and maxima both as a function of power and as a function of energy. In particular, strong gain is observed at different positions of the spectrum.

The experiments obviously agree with the model described in detail above. They are also very well reproduced by a more complete model: the solution of semiconductor Maxwell–Bloch equations at the Hartree–Fock level [31,53]. In particular, the strong oscillations around the exciton frequency, which are observed in the experiments, are reproduced by the calculations. They would not be obtained from the

solution of the Bloch equations described above for an ensemble of two level systems. However, the simple prediction of the occurrence of AC Stark bands, with a distance varying as the square root of the excitation power is well recovered.

## 7. Conclusions

To summarize, we have presented in the present paper a series of results evidencing the simple similarities of the excitonic non-linearities in quantum wells with atomic two-level systems. We have observed both field driven Rabi flopping and AC Stark splitting in different configurations. The first clear demonstration is obtained in the case of microcavities under strong coupling regime. In such systems, we also observe very strong hyper-Raman gain. We also observe directly the effects of strong coupling with the light field in a single quantum well by using a special configuration of the sample. These experiments show that, within some limits, the simple behavior of strong coupling in two-level systems may be reproduced in quantum wells. This demonstrates that, provided care is taken to limit the number of excited free electron-hole pairs, the dressing by the light field may be larger than the effects due to excitation induced dephasing. Excitonic correlations are therefore able to persist in the case of strong resonant excitation, and give rise to huge non-linearities.

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