

Direct observation of an ac Stark splitting in semiconductor microcavities excited above the continuum onset

F. Quochi, M. Saba, C. Ciuti, R. P. Stanley, R. Houdré, U. Oesterle, J. L. Staehli, and B. Deveaud
Physics Department, Swiss Federal Institute of Technology Lausanne, PH-Ecublens, CH-1015 Lausanne-EPFL, Switzerland

G. Bongiovanni and A. Mura

Dipartimento di Fisica and Istituto Nazionale di Fisica della Materia, Università degli Studi di Cagliari, I-09042 Monserrato (CA), Italy

(Received 8 November 1999)

We study the coherent optical response of a very high-finesse microcavity with a single quantum well by means of degenerate subpicosecond pump-probe spectroscopy. The dynamics of the ac Stark effect versus pump intensity is investigated upon tuning the empty-cavity mode below and above the exciton transition energy, at the exciton continuum onset. The evolution from a simple dynamical excitonic shift to a dynamical splitting is clearly observed in both cases. The qualitative agreement with numerical calculations based on the semiconductor Maxwell-Bloch equations is good.

Since the pioneering observation of the *excitonic ac Stark blueshift* in semiconductors,¹ many aspects of the transient coherent dynamics of photoexcited semiconductors have been understood by comparison with the physics of atomic systems. In the last decade, direct observations of a *Rabi flopping* from resonantly field-driven excitons²⁻⁴ have reinforced the establishment of a basic analogy between the coherent response of atoms and that of excitons in semiconductors.

A large body of experimental as well as theoretical works on the ac Stark effect in semiconductors, however, pointed out the influence of continuum states and related many-body effects.⁵

The role of Coulomb scattering has continuously been addressed. In the case of self-induced transparency from semiconductor excitons, excitation-induced dephasing (EID) has been found to decrease the transparency level and to dampen the Rabi flopping as well.³

As for resonantly excited III-V microcavity (MC)-embedded QW's, it was often believed that EID is the dominant nonlinearity in the excitation regime below the Mott density, which causes an abrupt collapse of the exciton-polariton Rabi splitting.⁶

Very recently, direct observation of a Rabi flopping and an *ac Stark splitting* of excitons has been reported in MC-embedded III-V QW's.⁷ Such an effect is the analog of the *Mollow spectrum* observed more than 20 years ago in sodium atomic beams,⁸ and clearly demonstrates by itself that EID is not the most important nonlinear effect of the coherent dynamics of resonantly pumped excitons in MC's. It has been found that *high-finesse* MC's excited by subpicosecond pulses are very suitable systems to observe a Rabi flopping and an ac Stark splitting in semiconductors.⁷

In fact, the cavity spectral filtering effect enables one: (i) to realize a high-resolution energy-selective excitation; (ii) to generate long-living field transients acting on the embedded semiconductor under examination while keeping an effective subpicosecond time resolution in measurements versus probe to pump delay.

A deep study of the transient coherent response of semiconductors embedded in high-finesse MC's is therefore important to establish a conclusive correspondence between the fundamental nonlinear processes in semiconductors and those in atomic systems.

In this paper, we report subpicosecond degenerate pump-probe experiments in a MC having an *extremely high finesse* and embedding a single QW.

Tuning the empty-cavity mode (i.e., the intracavity optical excitation) below and above the exciton transition energy, slightly above the continuum onset, we carefully study the ac Stark effect by analyzing the probe spectrum at zero time delay. Increasing the pump field intensity, the ac Stark effect evolves from a simple exciton line shift towards a dynamical splitting centered at the cavitylike resonance. Several field-driven Rabi oscillations are detected versus probe delay. The excitonic shift is toward *low* (high) energy for *positive* (negative) cavity to exciton detuning. The experimental results are compared with numerical solutions of the Maxwell-Bloch equations for semiconductors in the Hartree-Fock (HF) approximation. Calculations are done in a realistic pump-probe arrangement with subpicosecond input pulses. The qualitative agreement with the experimental results is good. This clearly demonstrates that selective excitation of low-kinetic-energy carriers near the continuum onset also permits the observation of the ac Stark effect in semiconductor QW's.

The investigated sample is grown by molecular-beam epitaxy on a GaAs substrate and consists of a single, 80-Å-wide $\text{In}_{0.03}\text{Ga}_{0.97}\text{As}$ QW positioned in the middle of a λ GaAs spacer. Top and bottom cavity mirrors are distributed Bragg reflectors made of 15 and 19 $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}/\text{AlAs}$ $\lambda/4$ pairs, respectively.

The linewidth of the empty-cavity mode is about $70 \mu\text{eV}$ (FWHM), which corresponds to a photon lifetime of 10 ps inside the empty cavity, and to a quality factor $Q > 2 \times 10^4$. The sample features strong exciton-photon coupling in the low-temperature and low-density limits. The ratio between the (low-field) Rabi splitting and the polariton linewidths is ≈ 30 (not shown). This means that the excitonic resonance is

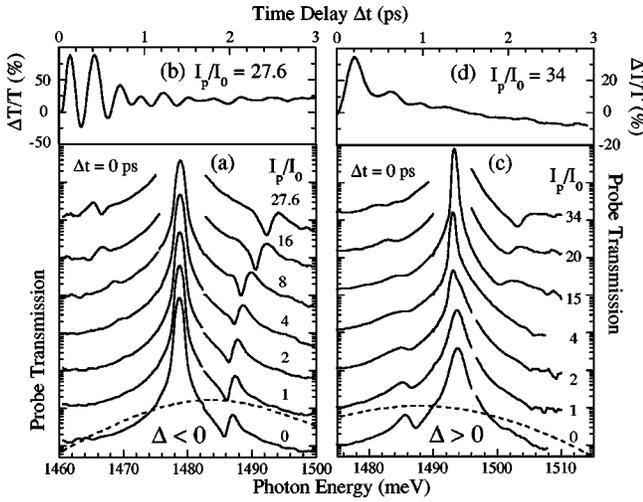


FIG. 1. (a) Probe transmission spectrum at zero probe delay ($\Delta t=0$), for different pump intensities. The cavity to exciton detuning is $\Delta=-7$ meV. Pump photon rates are given in units of $I_0=10^{13}$ photons cm^{-2} per pulse. The spectrum of the incident pump and probe pulses is plotted as dashed line. (c) Same as (a) but $\Delta=+7$ meV. (b) Probe transmission $\Delta T/T=(T(\Delta t)-T(0))/T(0)$, versus probe delay Δt . The cavity to exciton detuning is $\Delta=-7$ meV, and the pump intensity is $I_p=27.6 I_0$. (d) Same as (b) but $\Delta=+7$ meV and $I_p=34 I_0$.

almost homogeneously broadened.

The measurements are done with co-circularly polarized 100-fs pump and probe pulses, at near normal incidence on the sample. The transmitted probe light is spatially filtered to avoid effects of inhomogeneous excitation. Both the incident pump photon flux and the probe delay are varied. Pump intensities as large as 3×10^{14} photons cm^{-2} per pulse are used. The lattice temperature is 2 K.

Figure 1(a) shows the transmission spectrum of the probe pulses at zero probe delay for different pump intensities.⁹ Here the empty-cavity mode is tuned below the exciton mode. The cavity to exciton detuning Δ is equal to -7 meV. In this condition, the exciton and the cavity mode are only weakly coupled. The lower polariton practically coincides with the empty-cavity mode and allows us to excite the QW excitons mostly off resonantly, at low energy, while the input pulses spectrally cover both polariton resonances (dashed line). When increasing pump intensity, the excitonlike polariton starts to blueshift. This corresponds to the well-known excitonic Stark blueshift¹ which occurs in the perturbative excitation regime. At high pump fields, when the Rabi energy in the cavity becomes comparable to or even larger than the unperturbed cavity to exciton detuning, the probe spectrum changes into an almost symmetric ac Stark triplet.

The ac Stark triplet, previously reported in another sample for resonant excitation,⁷ occurs in the optical saturation regime for excitons. It represents the transient Mollow spectrum of ultrafast and coherently driven cavity-embedded semiconductors.¹⁰ In fact, we observe the evolution from a simple shift (perturbative excitation regime) towards a dynamical triplet with saturation of the excitonic resonance (nonperturbative excitation regime) when increasing the pump intensity.

Figure 1(b) depicts the temporal evolution of the probe transmission. The signal is acquired within a 0.7-meV-wide

band pass centered at the energy of the central peak of the triplet as a function of the probe delay. At positive delays, the trace features high-contrast field-driven oscillations.¹¹

The beating energy of the oscillations corresponds to the measured ac Stark splittings.⁷ As confirmed by a basic dynamical Maxwell-Bloch model,¹⁰ these oscillations correspond to a real Rabi flopping of the population created by the pump pulses in the QW, and are in fact the time-domain counterpart of the ac Stark splitting.

To investigate the possibility of observing an ac Stark effect degenerate with continuum states, we now tune the pump at energies *higher* than the exciton transition energy.¹² In Fig. 1(c), the probe transmission spectrum at zero delay is plotted versus pump intensity, for a cavity to exciton detuning $\Delta=+7$ meV.

Owing to the fact that in this $\text{In}_{0.03}\text{Ga}_{0.97}\text{As}$ QW the exciton binding energy is ≤ 5.5 meV,¹³ we actually realize an *energy-selective* excitation slightly above the continuum onset.

Coupling between the cavity and the continuum states results in a broadening of the upper (cavitylike) polariton [lowest curve of Fig. 1(c)]. Most of the observations concerning Fig. 1(a) apply also to Fig. 1(c).

Actually, the system response to the probe pulses evolves from an ac Stark *redshift* of the lower (excitonlike) polariton to an ac Stark triplet.¹⁴

Both ac Stark dips are smoother than in the case of low-energy excitation. Resonant pumping at the continuum onset results in fact in larger dephasing rates. This also affects the visibility of the field-driven Rabi oscillations detected versus probe delay [see Fig. 1(d)].

We notice that in both cases of low-energy [$\Delta \leq 0$, Fig. 1(a)] and high-energy [$\Delta > 0$, Fig. 1(c)] excitation the high-energy ac Stark dip is more pronounced than the low-energy one. The intensity asymmetry of the ac Stark splitting is not astonishing if one considers that a continuum of resonances lies above the “atomiclike” excitonic resonance.

We now analyze quantitatively the dynamics of the splitting occurring in the probe spectrum versus pump intensity. The Rabi flopping energy is estimated by measuring the splitting between the Stark dips and the cavity mode, as in Ref. 7. The energy separation between the high-energy (low-energy) dip and the cavitylike peak in the probe transmission is reported in Fig. 2(a) (b) as a function of the pump photon rate I_p (squares). As the ac Stark effect starts out of resonance with respect to the exciton transition, the Rabi flopping energy \mathcal{R} should have the following expression:

$$\mathcal{R} = \sqrt{\mathcal{R}_0^2 + \Delta^2}. \quad (1)$$

Here Δ is the zero-field cavity to exciton detuning and \mathcal{R}_0 is the HF Rabi energy $\mu_{cv}E_p +$ many-body terms (μ_{cv} is the interband dipole moment and $E_p \sim I_p^{1/2}$ is the intracavity electric field amplitude of the pump pulses). This fact is checked by plotting the quantity $\sqrt{\mathcal{R}^2 - \Delta^2}$ versus pump intensity (circles). A fit to the data shows that $\sqrt{\mathcal{R}^2 - \Delta^2}$ varies as $I_p^{1/2}$, approximately, in both cases of negative and positive exciton to cavity detunings. This leads us to infer that many-body corrections are either negligible with respect to $\mu_{cv}E_p$ (at high pump fields) or proportional to E_p itself.

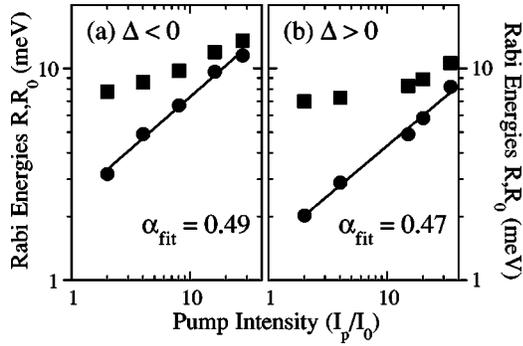


FIG. 2. Rabi energies \mathcal{R} (estimated as explained in the text, squares) and $\mathcal{R}_0 = \sqrt{\mathcal{R}^2 - \Delta^2}$ (circles) versus renormalized pump intensity $\mathcal{I} = I_p/I_0$. Δ is the cavity to exciton detuning and $I_0 = 10^{13}$ photons cm^{-2} per pulse. Continuous lines are fits to \mathcal{R}_0 using a power function $f(\mathcal{I}) \propto \mathcal{I}^\alpha$. Cavity to exciton detuning: (a) $\Delta = -7$ meV; (b) $\Delta = +7$ meV.

The observation of a Rabi flopping and a dynamical Stark splitting when exciting above the exciton continuum onset is a result of great importance. Such a result means that the phase-space filling nonlinearity can be more important than Coulomb interaction effects related to continuum absorption such as EID.

Actually, the further we tune the cavity mode and the laser pulses to high energies in the continuum region, the more the amplitude of the ac Stark dips decreases and for $\Delta \approx +10$ meV field-driven effects disappear (not shown). Vanishing of the ac Stark effect in the continuum is attributed to EID.

To evaluate the current way of interpreting our experimental results, we now present numerical calculations based on the semiconductor Maxwell-Bloch equations for a MC containing a QW and excited by 100-fs pump and probe pulses. The semiconductor Bloch equations (SBE) are treated at the level of the HF approximation in a two-band scheme, using a quasi-two-dimensional (2D) Coulomb potential.¹⁵

A SBE model is necessary for a theoretical investigation of the ac Stark effect in semiconductors excited above the fundamental excitonic resonance. Polarization scattering due to Coulomb interactions is taken into account phenomenologically, the collisional linewidth is taken to be proportional to the total density. To calculate the effective optical susceptibility of the cavity-embedded QW, the Maxwell equation is treated within quasimode approximation.¹⁶ The effective optical susceptibility is then inserted in a transfer-matrix calculation to obtain the optical transmission spectra of the weak probe pulses (see Ref. 10 again).

As for the parameters inherent to the calculations, we use an effective length of 200 Å for the quasi-2D Coulomb potential, which yields a realistic exciton binding energy of 5.3 meV (using the dielectric constant of GaAs). The zero-density line broadening is 1 meV. The probe transmission spectra are calculated at zero probe delay for various intensities of the applied pump field. Calculations are done: (a) For a cavity to exciton detuning of -7 meV with a collisional broadening rate of $0.25 \text{ meV} \times 10^{-10} \text{ cm}^2$; (b) for a cavity to exciton detuning of $+7$ meV with a rate of $0.75 \text{ meV} \times 10^{-10} \text{ cm}^2$. The field intensity lifetime in the empty

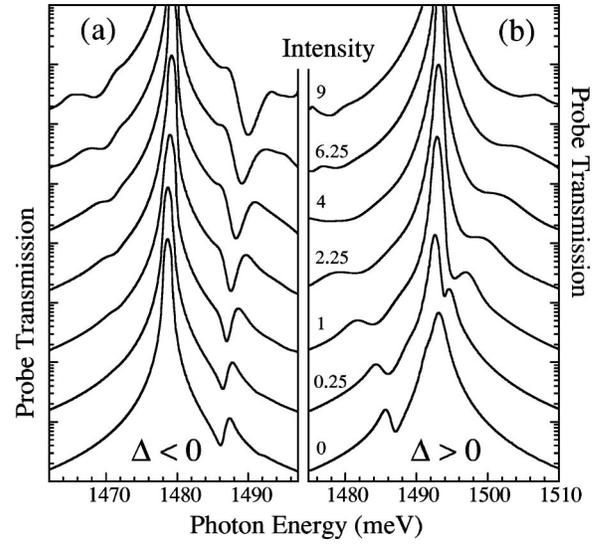


FIG. 3. Calculated probe transmission spectra at zero probe delay ($\Delta t = 0$) for different pump intensities. The intensity scale is in arbitrary units. (a) Cavity to exciton detuning: (a) $\Delta = -7$ meV; (b) $\Delta = +7$ meV. The parameters used for these calculations are reported in the text.

cavity is 10 ps, as experimentally observed, and the polariton coupling constant is adjusted for the observed zero-field Rabi splitting of 3.6 meV. It is worth noticing that the only relevant phenomenological parameter in our current calculations is the collisional broadening rate.

The theoretical results are reported in Fig. 3. Overall, the qualitative agreement with the experimental findings is remarkable. In both cases of negative and positive cavity to exciton detunings, a crossover occurs between the perturbative and the nonperturbative regime for the ac Stark effect. The first regime manifests itself just by a shift of the excitonlike polariton, while in the second one the spectrum features an ac Stark splitting around the (central) cavity mode.¹⁷ For $\Delta = +7$ meV, the initial exciton redshift (spectrum at $I = 0.25$, panel b) occurs simultaneously with the blueshift of the continuum onset, which causes the upper ac Stark sideband to appear on the high-energy tail of the cavitylike mode. Actually, the calculations overestimate the asymmetry of the ac Stark effect; this fact might be due to the lack of excitonic correlations in our HF model.¹⁸

Direct inspection of the temporal dynamics of the QW electric field, polarization, and population induced by a pump pulse confirms to a large extent the simple picture previously drawn on the basis of atomiclike single-resonance Maxwell-Bloch models.^{7,10} At sufficiently high pump rates, the pump pulse drives a coherent transient with a Rabi flopping of the QW population. When probing such a transient by means of a weak broadband pulse coherently coupled to the pump pulse, the pump-probe wave mixing causes an ac Stark splitting to occur in the probe spectrum.

As the pump field is spectrally filtered at the energy of the cavity mode, the dynamical splitting occurs around the cavity mode itself. At relatively high fields, the many-body (HF) corrections to the Rabi energy are found to increase linearly with the applied pump field, approximately.

This is consistent with the experimental finding $\mathcal{R}_0 \propto I_p^{1/2}$, and thus demonstrates that many-body effects do not size-

ably influence the qualitative phenomenology of the atomic ac Stark effect. Quantitatively, we find that at the highest pump intensities the HF many-body correction enhances the Rabi energy by $\approx 10\text{--}50\%$ (depending on time, wave vector, and detuning). Further, the role of EID is merely quantitative. Increasing the polarization scattering rate only results in a decrease of the amplitude of the Rabi flopping and of the ac Stark sidebands.¹⁹

In conclusion, we observe an ac Stark splitting in the optical spectrum of a MC-embedded QW when the system is excited by intense subpicosecond pulses in the exciton continuum. Numerical calculations based on the Maxwell-SBE

reproduce qualitatively the experimental results. Overall, our results suggest that Coulomb-induced dephasing due to optical absorption in the continuum region does not dominate the transient, nonlinear coherent dynamics of semiconductor QW's, as long as carriers are photogenerated with near-zero excess energy.

We gratefully acknowledge stimulating discussions with G. R. Hayes, C. Piermarocchi, and V. Savona. This work was partially supported by the Fonds National Suisse de la Recherche Scientifique, by the Programme Prioritaire OPTIQUE du Conseil des Ecoles Polytechniques Fédérales, and by the Italian Consiglio Nazionale delle Ricerche.

-
- ¹A. Mysyrowicz *et al.*, Phys. Rev. Lett. **56**, 2748 (1986); A. Von Lehmen *et al.*, Opt. Lett. **11**, 609 (1986).
- ²S.T. Cundiff *et al.*, Phys. Rev. Lett. **73**, 1178 (1994).
- ³H. Giessen *et al.*, Phys. Rev. Lett. **81**, 4260 (1998).
- ⁴A. Schülzgen *et al.*, Phys. Rev. Lett. **82**, 2346 (1999).
- ⁵See, e.g., W.H. Knox *et al.*, Phys. Rev. Lett. **62**, 1189 (1989); D.S. Chemla *et al.*, J. Lumin. **44**, 233 (1989).
- ⁶O. Lyngnes *et al.*, Solid State Commun. **104**, 297 (1997).
- ⁷F. Quochi *et al.*, Phys. Rev. Lett. **80**, 4733 (1998).
- ⁸F.Y. Wu *et al.*, Phys. Rev. Lett. **38**, 1077 (1977).
- ⁹These spectra are not representative of the very high quality of the sample, as in the current experiments the actual spectral resolution is limited to 0.7 meV.
- ¹⁰F. Quochi *et al.*, Phys. Rev. B **59**, R15 594 (1999).
- ¹¹At negative delays, owing to the interference between the polarization induced by the probe field and the pump field inside the QW, the transmitted probe signal features zero-field Rabi oscillations (not reported, see Ref. 7 again).
- ¹²We notice that for zero cavity to exciton detuning the results of Ref. 7 are completely reproduced.
- ¹³Tuning the cavity mode 5 meV above the exciton transition line provokes an abrupt and strong intensity decrease and broadening of the upper (cavitylike) polariton in the linear transmission spectrum. This fact is due to the resonant coupling between the cavity mode and the continuum band edge. A careful analysis of high-resolution linear reflection spectra versus cavity to exciton detuning allows us to put 5.5 meV as the upper limit for the binding energy of the fundamental QW exciton.
- ¹⁴Experimentally, it turns out that at the lowest excitation rates, resonant continuum absorption is less important than the off-resonant excitonic ac Stark effect.
- ¹⁵For a thorough review on the SBE, see H. Haug and S.W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 1990).
- ¹⁶H.J. Carmichael, in *Cavity Quantum Electrodynamics*, edited by P.R. Berman (Academic, Boston, 1994), p. 381.
- ¹⁷The pump-induced QW density grows with an increase in the applied pump intensity. At the maximum intensity the total density (measured after the first Rabi flop) is about 10^{11} cm^{-2} .
- ¹⁸This is a very interesting subject which certainly deserves further investigation.
- ¹⁹Accounting for polarization and population scattering within the field-dressed electron and hole energy bands at the second-Born level is found not to significantly alter the HF dynamics [C. Ciuti *et al.*, Phys. Rev. Lett. (to be published)].