Strongly Driven Semiconductor Microcavities: From the Polariton Doublet to an ac Stark Triplet

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We report femtosecond pump and probe experiments in a semiconductor microcavity containing quantum wells. At high pump fields, the exciton-polariton Rabi doublet changes into a triplet structure. The triplet splitting increases as the square root of the input intensity. The transmitted probe intensity is modulated with the corresponding frequency versus pump-probe delay. The experimental results are discussed in terms of Mollow spectra of a saturated transition of a two-level system. [S0031-9007(98)06194-8]

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Most of the studies reported on the strong exciton-photon coupling in semiconductor microcavities (MCs) [1] concern the regime of linear response [2]. The investigation of the excitonic nonlinearities under an intense optical field is also of great interest, especially when tuned at the energy of the exciton (X) and Fabry-Perot mode. The *resonant* nonlinear response is important for both applied purposes and fundamental physics. Potential applications of MCs concern optoelectronic devices, as, e.g., ultrafast all-optical switching, which should operate under strong and resonant excitation conditions [3]. From a fundamental point of view, previous studies concern nonlinearities due to a high density of carriers nonresonantly injected into the MC [4], while the number of available experimental and theoretical investigations in the high and resonant excitation regime is quite limited [5].

In semiconductors, the atomic two-level model has been widely used for modeling the coherent response of Xs [6], and even for X-MC systems [7]. In atomic physics, the two-level resonances have been extensively studied during the past decades. Mollow calculated both the free-space emission pattern (the so-called *Mollow triplet*) as well as the optical susceptibility (just called *Mollow spectrum*) of a strongly driven two-level system [8]. Fluorescence and absorption-gain measurements in atomic beams have verified these early theoretical predictions [9]. For the case of atoms inside a cavity that are *strongly* coupled to the electromagnetic field, to our knowledge the experimental results available concern only the normal-mode doublet and the Jaynes-Cummings dynamics observable at low fields [10]. With increasing field strength, theory predicts a transition from the Rabi doublet to an ac Stark triplet (or to even more complex spectra) [11].

The ac Stark effect of quantum well (QW) Xs, as it has been observed in pump-probe experiments, represents the analog of the Mollow absorption spectrum for a nonresonant low-energy pumping [12]. In principle, it should be possible to observe the resonant ac Stark splitting of Xs in high-quality samples at low temperatures, using ps laser pulses in order to avoid the excitation of continuum states which would make unstable the X resonance. However, such experiments have not yet been reported.

In this Letter, we show that a high-finesse semiconductor MC containing QWs is a much more suitable system. Our MC, featuring strong X-photon coupling, has been resonantly excited by fs laser pulses; the spectral filtering effect of the cavity prevents the excitation of dissociated electron-hole (e-h) pairs. To explore the coherent phenomena, the transmitted spectrum is probed by weak pulses at short delays. We observe, for the first time to our knowledge, the evolution from the normal-mode (polariton) doublet to an ac Stark *triplet*, when increasing the pump intensity. Correspondingly, the time-integrated transmitted probe intensity versus pump-probe delay features oscillations whose frequency increases as the inverse of the triplet splitting. The experimental observations are attributed to a dense coherent X population, near saturation. The spectra are qualitatively reproduced with a semiclassical model, solving the Maxwell wave equation with the original Mollow formula for the dielectric susceptibility of a strongly driven two-level system [8,13].

The investigated sample is a $3\lambda/2$ GaAs MC, containing six 75 Å wide $In_{0.13}Ga_{0.87}As$ QWs at the antinodes of the electric field. The cavity mode is resonant with the heavy-hole X state, where the exciton-photon coupling gives rise to a Rabi splitting $\hbar\Omega_0$ of 8 meV, at low densities. The sample features an inhomogeneous X broadening of about 5 meV, as previously reported [7]. 100 fs pulses were used at nearly normal incidence. The pump and probe pulses were crossed on the sample and arrived on the MC surface at $t = 0$ and $t = \Delta t$, respectively. Pump intensities up to 10^{14} photons/(pulse cm^2) were used. To minimize inhomogeneous spatial excitation, only the central part of the pump spot on the sample was probed. The experiments were done at a temperature of 2 K. In the weak excitation regime, transmission and reflection experiments show that $\approx 3\%$ of the incoming photons are absorbed by each quantum well in the MC. Assuming a saturation density $n_s \simeq 5 \times 10^{10}$ cm^{-2} (for a pure X gas [14]), we define a corresponding "saturation intensity" (measured outside the MC) $I_s = 2 \times 10^{12}$ photons/(pulse cm^2).

A sequence of transmission spectra, measured for a probe delay $\Delta t = 0$ and for different pump intensities, is reported in Fig. 1(a). Pump and probe are co-circularly polarized. Without pump, the probe spectrum shows the normal polariton splitting. For pump intensities slightly larger than I_s a narrow structure appears between the two polariton lines [15]. With increasing intensity, this peak becomes larger and shifts towards the frequency of the empty MC mode. On both sides of the central peak, dips can be observed. The distance between them increases with the pump field, which is typical for a dynamical Stark splitting. The level of the pump absorption, estimated from the measurements of both the transmission and reflection spectra, drops from the initial (low-intensity) value of about 3% to 0% (within the relative large experimental uncertainties) at the highest intensities, denoting a clear saturation behavior. A direct evaluation of the X density created by a pump pulse is, however, not possible.

The dynamics of the transmission spectrum is monitored for different probe delays in Fig. 1(b), for $I_p = 20 \times 10^{12}$ photons/(pulse cm^2). For short negative probe delays the central peak appears, and the lateral dips develop. Their spectral separation increases as Δt approaches 0, since they are dynamically induced by the strong electric field of the pump. The low-energy dip quickly disappears, and at $\Delta t \simeq 1$ ps (once the coherence of the system is lost) we observe again a polariton doublet. This doublet is, indeed, highly asymmetric, as the lower peak is drastically enhanced. However, the upper peak, which is spectrally unshifted, is not much weaker than the unperturbed one (even at the highest intensities). A simple analysis that will be reported elsewhere shows that the high asymmetry in the probe spectra is due to bleaching of the low-energy states in the inhomogeneous X distribution. Nevertheless, the high-energy excitonic levels are still strongly coupled to the cavity mode, giving rise to the observed polariton doublet. In other words, within the population created by the pump pulse the excitonic correlation is still preserved.

The evolution from the coherent to the incoherent regime can be monitored by looking at the transmitted probe intensity versus pump-probe delay. The intensity has been measured with a spectral bandpass of 1.3 meV at the energy of the central peak for $\Delta t = 0$. The signal trace is reported for different pumping levels in Fig. 2. The transmitted intensity reveals oscillations for both negative and positive delays. For $\Delta t \geq 0$ the period of the beatings decreases from 0.5 (the low-field Rabi period) to 0.2 ps with increasing excitation intensity.

A more quantitative analysis has been performed comparing the beating energies $E_{\mathrm{beat}} = h/T$ (calculated from

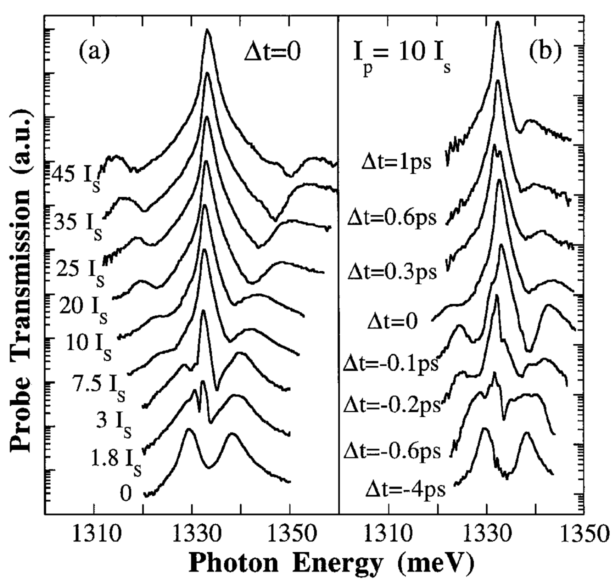

FIG. 1. Transmission spectra of the excited MC, as measured by the weak probe at a delay $\Delta t = 0$ for different excitation intensities I_p (a) and at $I_p = 10 I_s$ for different delays Δt (b). $I_s = 2 \times 10^{12}$ photons/(pulse cm^2).

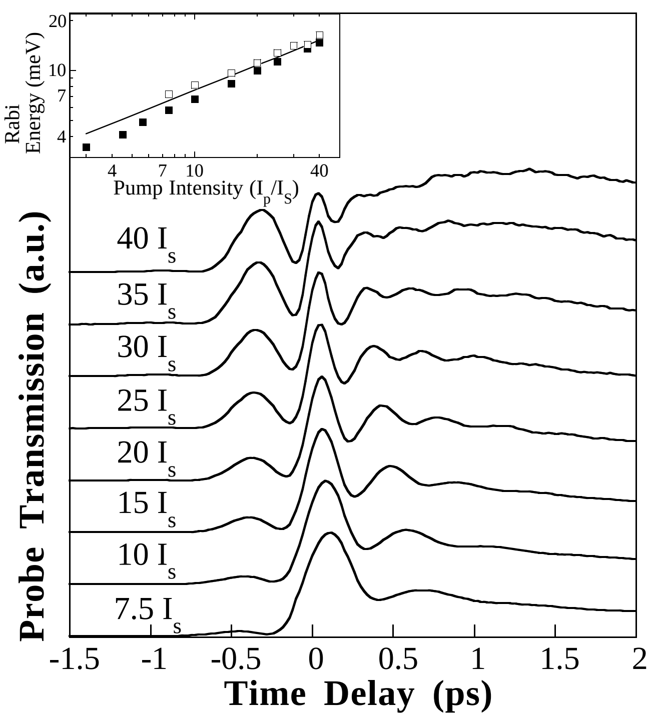

FIG. 2. Intensity of the probe pulses transmitted through the pumped MC versus pump-probe delay Δt, for different excitation densities. $I_s = 2 \times 10^{12}$ photons/(pulse cm^2). Inset: open squares, beating energies (E_{beat}), calculated as explained in the text; solid squares, half the separation energy ($\Delta E_{\mathrm{dip}}/2$) of the two lateral dips in the transmission spectra. Continuous line: $f(I_p) = \mathrm{const} \times I_p^{1/2}$.

the temporal intervals T between the first two maxima at $\Delta t \geq 0$) with half the dip splittings $\Delta E_{\text{dip}}/2$. The corresponding values have been reported as a function of I_p in the inset of Fig. 2. They are very similar and scale as $I_p^{1/2}$. Oscillations are damped, due to dephasing processes. Nevertheless, the oscillation decay time does not decrease significantly with the pump level, not more than a factor of 3 (a quantitative evaluation is not trivial). Together with the saturation behavior of the absorption, this fact suggests that the X density does not exceed n_s [16]. As a consequence, we infer that both the triplet and the fast oscillations come from the coherent response of a dense X population to a very intense intracavity electric field, without a sizeable population of dissociated e-h pairs. The saturation of the pump absorption allows large intracavity intensities to be attained during the excitation, as required to observe such field-driven phenomena.

In order to explain the previous experimental results, we consider the Mollow optical susceptibility χ for a weak probe. The analytical expression (valid in the case of a monochromatic pump wave [13]) is the following:

$$\chi(\omega, \omega_p, \omega_x) \equiv \chi_{\text{inc}} + \chi_{\text{coh}} = \frac{\hbar \Gamma_{\text{rad}} D(\omega)}{1 + \gamma I_p \operatorname{Re} D(\omega_p)} \times \left(1 - \frac{1/2 I_p F(\omega_p - \omega)[D(\omega) + D^*(\omega_p)]}{1 + 1/2 I_p F(\omega_p - \omega)[D(\omega) + D^*(2\omega_p - \omega)]}\right). \tag{1}$$

Here, ω, ω_p, and ω_x are the frequencies of the probe, pump, and unperturbed X transition, respectively; Γ_{rad} is the radiative decay rate of the unsaturated X transition; $I_p \equiv \Omega_p^2/(\gamma\Gamma)$ is a dimensionless pump intensity; $\hbar\Omega_p \equiv \mu E_p$ is the Rabi energy (of the Xs in the resonant pump field E_p); γ and Γ are the transverse and longitudinal relaxation rates; $D(\omega_i) \equiv \gamma/[\gamma + i(\omega_x - \omega_i)]$, and $F(\omega_p - \omega) \equiv \Gamma/[\Gamma + i(\omega_p - \omega)]$. Equation (1) contains all orders in the pump field, and the first order in the probe field. The term χ_{coh} is due to a process of diffraction of the pump off the polarization grating induced by the interfering pump and probe fields: It thus represents the coherent pump-probe coupling. For $I_p \to 0$, χ_{coh} vanishes, and χ_{inc} becomes the usual linear susceptibility.

The approximation of the monochromatic pump wave can seem very crude. However, it is worth mentioning that for $I_p \gg I_s$, when strong X bleaching occurs, the pump pulses are reshaped by the cavity filtering effect, which drastically reduces their spectral width ($\simeq$15 meV) to $\simeq$1 meV (i.e., the linewidth of the empty cavity mode, not reported). This value is considerably smaller than both the polariton splitting $\hbar\Omega_0$ and the inhomogeneous X linewidth, and comparable to the homogeneous one, given by the damping time of the oscillations.

The transmission of a weak pulse can be calculated solving the Maxwell wave equation with the susceptibility of the QWs given by Eq. (1). This is done by means of a standard transfer matrix method for the boundary conditions of the probe electric field [17]. The obtained probe transmission spectra, together with the free QW absorption coefficient $\alpha(\omega) \propto \omega \operatorname{Im}[\chi(\omega)]$, are shown in Figs. 3(a) and 3(b), respectively, for different Rabi energies $\hbar\Omega_p$. For simplicity, we have assumed all parameters to be intensity independent: (i) $\omega_p = \omega_x = \omega_{\text{cav}}$ ($\hbar\omega_{\text{cav}}$ is the empty cavity energy); (ii) $1/\Gamma = 1/2\gamma = 1.5$ ps. The inhomogeneous broadening of the X resonance has been included by assuming that the susceptibility is given by $\int G(\omega_x)\chi(\omega, \omega_p, \omega_x)\, d\omega_x$, where $G(\omega_x)$ is a Gaussian with a width of 5.5 meV (FWHM) [7]. The behavior of the calculated transmission spectra versus pump intensity reproduces the salient features of the experimental observations. Increasing $\hbar\Omega_p$, the transmission at the pump laser frequency becomes dominant. The central peak is due to the saturation of the two-level system at $\omega = \omega_p$ where α approaches zero. The splitting between the lateral structures, namely, the dynamical Stark splitting of the Mollow spectrum, comes from χ_{coh}. It depends only on $\hbar\Omega_p$ and it scales as $\hbar\Omega_p \propto I_p^{1/2}$. At sufficiently high intensities, the hole burnt by the pump into α becomes wider and two lobes with slightly negative α appear. For $\omega - \omega_p \simeq \pm\Omega_p$, α is near its maximum value, causing the two sidewings in the transmission spectra [18].

The computed spectra yield an estimate of the strength of the coupling between the field and the system: The experimental values $\Delta E_{\text{dip}}/2$ can be taken as measurements of the Rabi energy in the cavity. An alternative and more

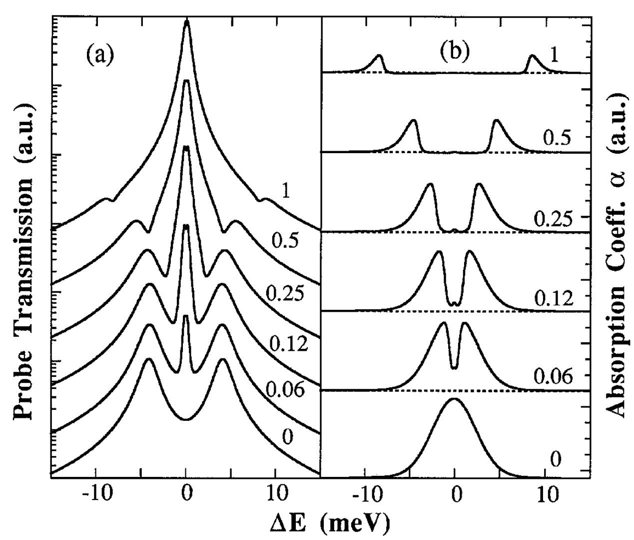

FIG. 3. (a) Transmission of the pumped MC, and (b) absorption coefficient of the excited QWs versus probe to exciton (X) detuning $\Delta E \equiv \hbar(\omega - \omega_X)$. The different values of the Rabi frequency Ω_p are given on the right in units of the low-field Rabi frequency Ω_0. Horizontal dashed lines mark zero absorption. The spectra have been calculated as described in the text.

direct estimate can be given. For an external input intensity $I_p \simeq 10^{14}$ photons/(pulse cm^2), we measured a pump transmission of about 3.5%. Solving the Maxwell wave equation [19], we find the intensity of the intracavity pump pulse to be $\simeq 1.5 \times 10^2$ times higher than the transmitted one. Finally, using a Kane energy value of 25 eV (as for bulk GaAs [20]) to evaluate μ, we obtain $\mu E_p \simeq 15$ meV inside the cavity, in very good agreement with our experimental results.

We add some observations of a more general character, concerning the possibility of observing such phenomena in different environments for Xs (free QWs and MCs). In an experiment with pulsed excitation, several favorable conditions are required to observe coherent oscillations related to the dynamical Stark effect. The duration of the excitation has to be longer than the Rabi period, and adequate temporal resolution has to be achieved. In free QWs these conditions cannot be satisfied simultaneously, because the temporal length of the laser pulses gives the excitation duration and the resolution, at the same time. Further, the use of ultrashort pulses resonant with the unperturbed X transition causes the simultaneous excitation of large numbers of continuum e-h pairs, making the X resonance unstable. In a MC, approaching the X saturation, the resonant femtosecond pulses are reshaped due to the cavity filtering: Inside our cavity they have a single-sided exponential shape (highly asymmetric), with a decay time $\simeq 1$ ps. The rise time is still given by the extracavity duration (~ 100 fs), approximately, allowing subpicosecond pump-probe timing to be kept. The spectral narrowing drastically reduces the excitation of continuum states as well. The other few works on experiments with femtosecond resonant excitation in MCs do not report the observation of the ac Stark splitting, they just mention the "collapse" of the two polariton peaks into a single one [5]. In these experiments, the quality factor of the empty cavity was about 4 times lower than ours, i.e., the cavity linewidth was of the same order of magnitude as the X binding energy, a situation which is not too far from that of free QWs [21]. Besides, it is worth remarking that in the experiments of Ref. [5] the measurements were performed in backgeometry. In this configuration, the weak spectral dips are hardly observable, as the spectra are dominated by the laser reflection.

In conclusion, this paper provides experimental evidence for the evolution of the low-field polariton doublet of a semiconductor MC into an ac Stark triplet and for the corresponding increase of the X Rabi frequency when an increasing resonant femtosecond excitation is applied. The basic experimental findings are attributed to the saturation of the Xs in the MC. Because of the strong filtering effect, we believe that a high-finesse semiconductor MC containing QWs, excited by ultrafast laser pulses, is relatively well suited to observe such phenomena.

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