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Strong coherent gain from semiconductor microcavities in the regime of excitonic saturation

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We present femtosecond gain measurements in microcavity-embedded quantum wells. When the excitonic transition is saturated by an intense pump field, the spectrum measured with a weak probe pulse is strongly modulated by pump-probe wave mixing processes. A giant probe amplification occurs within the bandwidth of the empty-cavity mode. The optical feedback provided by the microcavity is responsible for a net gain of the broadband probe pulse. [S0163-1829(99)50424-4]

The nonlinear optical response of excitons strongly and coherently driven by intense laser pulses is a topic raising much interest.[1] So far, femtosecond (fs) pump-probe experiments in free-space semiconductors, with the pump pulses tuned to the transparency region below the band gap, showed the so-called *optical Stark shift* of the exciton resonance.[2] Similar experimental investigations have recently reported a *hyper-Raman gain* on the low-energy side of the nonresonant pump pulses.[3] These phenomena, involving complex absorption/gain patterns in the spectum of the outgoing probe pulses, originate from coherent pump-probe wave mixing processes which are significant for the comprehension of fundamental physics of coherent excitation in semiconductors. The corresponding amplification is often referred to as *coherent gain*. However, in free-space semiconductors one deals with very small coherent gain, revealed only for intensities not exceeding the limit of exciton optical saturation.[2] In a semiconductor microcavity (MC), probe stimulated emission occurring within the bandpass of the empty-cavity mode is expected to undergo an enhancement related to the cavity quality factor Q. The choice of optimized Q factors and probe pulse widths would also bring MC's closer to requirements for practical applications in ultrafast optical devices.

The intriguing case of a saturating *resonant* excitation in MC's containing quantum wells (QW's) has been addressed very recently.[4–6] In a high-finesse cavity, it has been shown that the exciton polariton doublet changes into an *exciton Stark triplet* when the system is driven by intense fs laser pulses.[6] The main experimental findings have been traced back to the fundamental properties of an optically saturated two-level system (2LS). This result, surprisingly simple if one considers the many-body nature of a strongly excited semiconductor, has been confirmed in a recent theoretical work based on the semiconductor Bloch equations (SBE), provided that the influence of continuum states on the system polarization is negligible.[7] The symmetric Stark sidebands are characteristic features of the regime of coherent saturation of the exciton transition, and quite remarkably are visible even at pumping rates I_p exceeding by far the exciton saturation intensity I_s.[8] Therefore, a high-finesse MC appears to be the perfect laboratory to study coherent gain phenomena in such a nonlinear regime. This represents a new challenging field in semiconductors.

In this paper, we investigate the coherent gain dynamics in a MC having $Q \simeq 2\times 10^3$, by means of fs degenerate and doubly resonant pump-probe experiments (i.e., the laser pulses are resonant with the cavity mode and the fundamental QW exciton). In the exciton saturation regime, the probe light is amplified up to more than a factor of 2 close to the energy of the empty-cavity mode, where positive optical feedback is achieved. Moreover, the spectrally integrated response shows a *net* gain of about 10%. Temporally, the gain is revealed at very short pump-probe delays and is characterized by strong modulations. The experimental results are compared to numerical solutions of the Maxwell-Bloch equations for an ensemble of 2LS's. The calculations give a qualitative explanation of the gain phenomenology in terms of pump-probe *coherent* coupling in optical cavities.

The experiments have been performed on a $3\lambda/2$ GaAs MC containing six 75 Å wide $In_{0.13}Ga_{0.87}As$ QW's placed at the antinodes of the electric field. When the cavity mode is resonant with the heavy-hole exciton resonance, the Rabi splitting Ω_R is $\simeq 8$ meV. The exciton inhomogeneous broadening and the linewidth of the empty cavity amount to about 5 meV and 0.7 meV, respectively. The MC, held at a temperature of 2 K, has been resonantly excited by circularly polarized 100 fs pulses with intensities I_p up to 7×10^{13} photons pulse^{-1} cm^{-2}, i.e., 35 times the exciton saturation intensity $I_s = 2\times 10^{12}$ photons pulse^{-1} cm^{-2}.[6,9] The response of the MC has been probed by means of copolarized pulses of much weaker intensity, at different delays Δt. To study the process of light amplification in the MC, the sum of the intensities of the reflected and the transmitted outgoing probe pulses, $\mathcal{R}\cdot I_t + \mathcal{T}\cdot I_t$, should be measured ($\mathcal{R}$ and $\mathcal{T}$ are the reflectivity and transmittivity, respectively, and I_t is the weak intensity of the incident probe). However, in our sample the transmitted intensity is found to be negligible with respect to

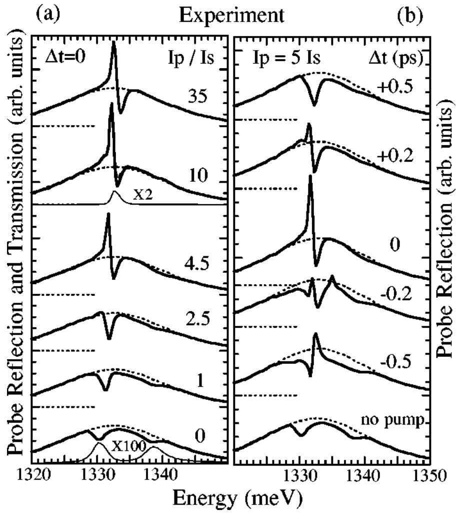

FIG. 1. Reflection spectra of the weak probe pulses: (a) for different pump intensities (I_p) at zero pump-probe delay (Δt); (b) for different Δt values at $I_p = 5I_s$ ($I_s = 2\times10^{12}$ photons pulse^{-1} cm^{-2}). The bold dashed curve is the sum of transmitted and reflected probe intensities at $I_p = 10I_s$. The smooth dashed curves represent the spectra of the incident probe pulses, while the horizontal dashed lines mark zero reflection. Two transmission spectra, at $I_p = 0$ and $10I_s$, are plotted as thin solid lines, with a relative magnification of 100 and 2, respectively.

the reflected one. This is due to the fact that the transmittivity of the top Bragg reflector (1.5%) is much higher than that of the back reflector (0.2%).

The main experimental results are presented in Fig. 1. Panel (a) shows a series of probe reflection spectra $\mathcal{R}\cdot I_t$, taken at zero pump-probe delay for increasing pump intensities. The spectra of the incident probe pulses (smooth dashed curves) serve as baselines to determine the fraction of amplified light $\mathcal{R}\cdot I_t - I_t$. At low intensities, the linear response gives rise to the polariton doublet. Increasing the pump intensity, the MC spectra drastically change. In *transmission*, the low-field polariton spectrum changes into the ac Stark triplet, and the energy of the central peak, essentially due to saturation of the exciton resonance, approaches that of the empty-cavity mode. A spectrum is plotted for $I_p = 2\times10^{13}$ photons pulse^{-1} cm^{-2}, upper thin solid line in panel (a). Due to the cavity spectral filtering, the ac Stark sidebands are very weak, and are not visible in a linear scale. Also, in the *reflection* spectra the most important changes are occurring close to the empty-cavity mode. For $I_p \approx 10^{12}$ photons pulse^{-1} cm^{-2}, the lower polariton dip becomes dominant, and with increasing pump intensity it shifts to the blue towards the center of the spectrum, as the exciton-photon coupling decreases. Once I_p approaches 10^{13} photons pulse^{-1} cm^{-2}, the dip evolves to a quite sharp dispersive-like structure featuring strong optical gain. In a narrow spectral band on the left part of the structure, the reflected intensity largely exceeds the incident one. This *giant* gain peak reaches values higher than 100% (i.e., $\mathcal{R} > 2$), and *persists* up to the highest pump intensities ($I_p \gg I_s$), when the exciton transition is strongly bleached.[10]

To show the irrelevance of the transmitted intensity for the estimate of the gain, the sum spectrum of transmitted plus reflected probe intensities ($\mathcal{R}\cdot I_t + \mathcal{T}\cdot I_t$) is plotted for $I_p = 2\times10^{13}$ photons pulse^{-1} cm^{-2} (bold dashed curve). As a matter of fact, it is almost indistinguishable from that of the reflected intensity $\mathcal{R}\cdot I_t$ alone. Importantly, the intensity of the spectrally integrated total probe emission exceeds the incident one by $\simeq 8\%$. We therefore infer that the strong cavity feedback also ensures *net* gain of our broadband probe pulses.[11]

In the reflection spectra it is even more difficult to reveal the ac Stark sidebands than in transmission, because outside the bandwidth of the empty-cavity mode $\mathcal{R}$ is $\simeq 1$. High-sensitivity differential measurements (not reported) have been necessary to distinguish them from the reflected laser intensity.

As shown in Fig. 1(b), close to zero delay the measured reflection spectrum depends sensitively on Δt. As a result of the coherent interaction between the tail of the polariton wave created by the weak probe and the strong pump field, a gain peak appears already at negative delays.

The sharp S-shaped structure is strongest at $\Delta t \simeq 0$. At $\Delta t \simeq +0.5$ ps it has already vanished and incoherent saturation of the exciton resonance appears. Actually, for this pump intensity the system has not yet reached the regime of strong saturation. Once a strong ac Stark effect takes place ($I_p \approx 1.5 - 2\times10^{13}$ photons pulse^{-1} cm^{-2}), the amplified probe intensity oscillates versus delay with a frequency corresponding to half the splitting of the Stark triplet, as already reported for the transmitted probe intensity.[6]

To model the pump-probe coherent processes, we adopt a two-level approximation for the fundamental exciton resonance. The interaction between the excitons and the intracavity pump and probe fields is described by coupling the optical Bloch equations for the 2LS to the Maxwell's wave equation. Our aim is to solve them numerically in a time-resolved case with fs incident pulses. For simplicity, we make a quasimode approximation for Maxwell's wave equation.[12] In order to account for the many-body nonlinearities of excitons, both the coupling factor g between the cavity mode and the 2LS and its optical transition energy E_X (the 1s exciton energy) are renormalized phenomenologically following the results of the perturbative theory developed by Schmitt-Rink *et al.*[9] This way, local-field corrections to the system Rabi energy[13] are automatically included. Further, accounting for a density-dependent exciton collision broadening is essential in a realistic description of the temporal dynamics[14]. This broadening is assumed to be linearly dependent on the exciton density. The exciton inhomogeneous broadening is accounted for by using a collection of 2LS's, spread in energy with a Gaussian distribution.

The calculation of the optical response of the MC system follows a two-step scheme. First, to include self-consistently the cavity field dynamics, we solve numerically the coupled Maxwell-Bloch equations and derive the linear optical susceptibility $\chi^{(1)}(\omega,\Delta t)$ for the weak probe, coherently coupled to the strong pump. Secondly, the susceptibility is inserted in a transfer matrix calculation to obtain the MC optical spectra. Details of the model will be reported in a forthcoming paper.

In Fig. 2, we compare experimental transmission spectra at zero delay,[6] panel (a), to calculations of transmission spectra, panel (b), and to the corresponding spectra of the QW absorption coefficient $\alpha(\omega,0) \propto Im[\chi^{(1)}(\omega,0)]$, panel (c). In

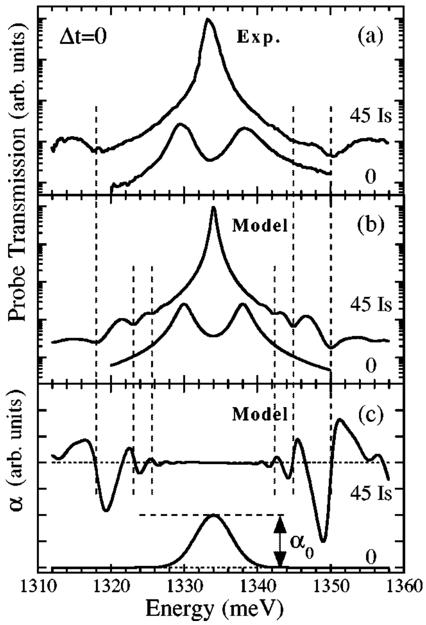

FIG. 2. (a) Measured probe transmission spectra at zero delay for $I_p=0$ and $I_p=45I_s$ ($I_s=2\times10^{12}$ photons pulse^{-1} cm^{-2}). (b) Calculated transmission spectra for the same cases as in (a). (c) Corresponding theoretical results for the absorption coefficient α (α_0 is the peak value at zero field). The vertical dashed lines mark the spectral positions of the Stark sidebands, while the horizontal dotted ones in panel (c) are reference zeros. Parameters are given in the text.

all panels, the lower and upper curves refer to the cases of zero and very high pump intensity ($I_p=9\times10^{13}$ photons pulse^{-1} cm^{-2}), respectively. Our model reproduces well the ac Stark effect of the resonantly saturated excitons inside the MC. In the α spectrum, the Gaussian line shape of the linear regime evolves, at strong pump intensities, towards bleaching of the optical transition close to the center of the spectrum and strong absorption-gain Stark sidewings.[15] Due to the finite temporal length of the excitation, in addition to the two main structures, minor ones occur. These features faintly appear also in the experimental spectra. Each band crosses zero absorption and causes a dip to appear in the MC transmission spectrum. However, the filtering effect of the cavity drastically reduces the strong gain lobes occurring far from the energy of the cavity mode. The slight asymmetry of the calculated spectra, in agreement with the experiment, is due to the exciton blueshift. We point out that in our model the only adjustable parameters are the exciton blueshift and the homogeneous broadening at saturation. In all simulations we have taken the values of 3 meV (Ref. 16) and 5 meV, respectively.

Now, let us address the strong coherent gain appearing in reflection. We first discuss a series of calculated reflection spectra of the probe, obtained by varying the pump intensity at $\Delta t=0$, Fig. 3(a). When the pump level is raised, the Rabi splitting starts collapsing. The asymmetry of the bleaching process comes from the interplay between renormalization of g and the exciton blueshift. Note that, in the simulations, the effective saturation intensity is lower than the experimental one by nearly a factor of 5. This can be imputed to the quantitative inadequacy of our basic modeling. For a reliable theoretical estimate, the density dependence of the exciton

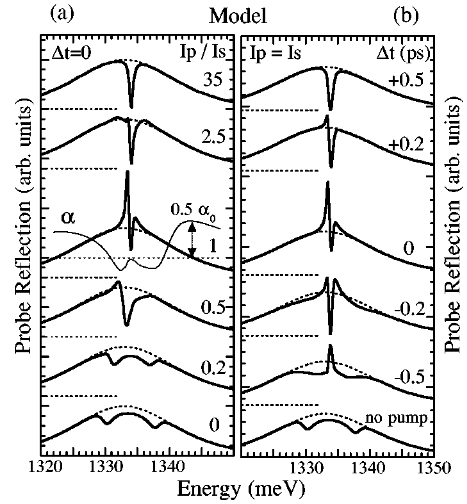

FIG. 3. Calculated reflection spectra of the probe: (a) at $\Delta t=0$ for different pump intensities I_p; (b) at $I_p=I_s$ for various delays. Horizontal and smooth dashed lines as in Fig. 1. The thin solid line in panel (a) is the spectrum of the absorption coefficient α for $I_p=I_s$ and $\Delta t=0$. The α scale is reported in units of α_0, defined in Fig. 2.

oscillator strength and linewidth should be properly calculated using reliable treatments of many-body interactions.

Increasing further the excitation level, coherent effects start taking place in the optical susceptibility. At $I_p=I_s$, the QW absorption coefficient α already shows Stark sidebands [see thin solid line in Fig. 3(a)]. Again, due to the blueshift of the resonance, the spectrum is asymmetric. The negative lobe at low energy, closer to the energy of the cavity mode than the high-energy one, is amplified by the cavity effect leading to the strong gain peak in the reflected light ($\mathcal{R}\simeq2$). Such resonant amplification is so large that a net gain still occurs in the spectrally integrated response ($\simeq4\%$ at $I_p=I_s$). Augmenting further the pump field, the Stark sidebands move rapidly out of the empty-cavity bandwidth and hence (resonant) gain *disappears*. This is the intrinsic phenomenology of the ac Stark effect for a 2LS (and for a SBE model restricted to the 1s-exciton resonance[7]). The persistence of the gain in the experimental spectra [Fig. 1(a)] strongly suggests that the off-resonantly excited *e-h* pair states also contribute to the coherent response with gain modulations at the energy of the cavity mode. This topic deserves a particular attention for all potential implications. For a proper theoretical treatment, the many-body approach of Ref. 7 should be extended to both excited exciton and continuum states at the band edge.

The spectra of Fig. 3(b) have been calculated at $I_p=I_s$, for which calculations foresee an incipient ac Stark effect at $\Delta t=0$, rather equivalent to the experimental series of Fig. 1(b). Probe amplification already occurs at $\Delta t<0$. At negative delays, the interference between the tail of the probe polarization wave with the saturating pump field causes spectral modulations to appear in the probe QW response. Amplitude and phase of these modulations depend sensitively upon pump-probe delay, resulting in a complex transient dynamics, strongly enhanced within the cavity mode bandwidth.[17] Such spectral modulations are temporal *precursors* of the ac Stark effect,[18] and, therefore, are expected to occur even for strong saturation. Actually, transient features for $\Delta t<0$ do not change qualitatively for $I_p \gg I_s$, in agreement with the experimental findings (not reported). They are characteristic features of the entire temporal dynamics of the ac Stark effect of excitons.

In conclusion, we report doubly resonant fs gain experiments on MC-embedded QW's. Coherent effects, occurring in the regime of the optical saturation of the exciton transition, lead to very sharp transient modulations in the spectra of weak probe pulses. The Maxwell-Bloch equations for 2LS's have been used to model in a basic way the pump-probe coherent coupling in strongly excited MC's. This basic approach is able to describe the transient ac Stark splitting of excitons, as well as its precursors (transient spectral modulations at negative delays). We demonstrate, experimentally and theoretically, that nonlinear pump-probe mixing in microcavities can produce giant coherent gain in resonance with the cavity mode as well as net coherent gain of a broadband probe pulse.

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[17] Note the oscillatory behavior (versus Δt) of the reflectivity $\mathcal{R}$ in the central part of the spectrum by comparing the two spectra at $\Delta t=-0.5$ and -0.2 ps.

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