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Direct observation of the excitonic ac Stark splitting in a quantum well

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Ultrafast pump-probe measurements evidence the dynamical Stark splitting in the exciton absorption spectrum of a single semiconductor quantum well. The intensity of the Stark sidebands in the spectrum is comparable to the linear absorption and their separation scales as the pump field amplitude. In the time domain the Rabi oscillations are observed. The semiconductor Bloch equations solved in the Hartree-Fock approximation demonstrate that the coherent exciton-light coupling dominates over the many-body dephasing.

The optical Stark effect originates from the nonlinear coupling between a strong oscillating electromagnetic field and an optically saturable electronic transition, e.g., between two atomic levels.[1,2] The strength of the interaction is called the Rabi energy. If the optical field is detuned in energy with respect to the electronic transition (off-resonant or perturbative effect), the Stark effect appears as a shift of the optical transition energy. When instead the Rabi energy exceeds the detuning (resonant or nonperturbative effect), then the external electric field is no more a perturbation to the electronic system but actually mixes its quantum eigenstates and splits into two sidebands the original absorption line.[3] In the time domain the population oscillates (Rabi oscillations) between the two sidebands, which are the eigenstates of the electronic system ''dressed'' by the electromagnetic field.

The excitonic transition in semiconductors is optically saturable and therefore can couple nonlinearly to the electric field.[4] But compared to the atomic systems, the dephasing of the electronic system (from both homogeneous and inhomogeneous sources) is much faster, so that also the coherent exciton-light coupling is rapidly destroyed. Very intense fields, achieved by means of pulsed laser excitation, were therefore necessary to observe the excitonic ac Stark shift (the perturbative effect).[5,6] A peculiarity of excitons is that they strongly interact because of the Coulomb force, giving rise to a nonlinearity not observed in atoms. Many studies have addressed the very rich phenomenology generated by the interplay of Stark effect and Coulomb nonlinearity.[7–14] One prominent interaction effect is the augmentation of the dephasing rate of the electronic states for growing population densities (collision broadening or excitation-induced dephasing). Therefore, the investigation of the resonant effect is problematic: if the optical field is set to the resonance with the transition, it excites a large population and the excitation-induced dephasing becomes faster and faster. Whether the nonperturbative Stark effect is observable or not depends on which nonlinearity dominates the semiconductor optical response for high excitation regimes, the nonlinear exciton-light coupling or the many-body interaction.

The evidence for the resonant ac Stark effect in semiconductors has been recently reported. The splitting has been observed in microcavities, where the incoming laser pulses are filtered by the Fabry-Pérot resonator and an intense electric field is achieved without the excitation of free electron-hole pairs.[15] But the ac Stark sidebands are necessarily detuned far away the cavity spectral filter and their intensity in the transmission spectrum is highly suppressed. The Rabi oscillations of the population driven by the pump field were also reported. Several Rabi oscillations have been observed also in free-space quantum wells by selectively exciting the heavy-hole exciton transition and detecting the effect of the oscillations at the light-hole exciton energy.[16] Achieving the nonperturbative optical Stark effect is therefore possible. However, the direct spectral observation of the ac Stark splitting of free-space excitons is still lacking.

In this paper we present the evolution of the excitonic absorption line of a single quantum well with growing resonant laser pumping. The sample is specially designed to artificially enhance the coupling of the quantum-well excitonic transition with the light, permitting us to directly measure the absorption spectrum with a high signal-to-noise ratio. The nonperturbative regime of the ac Stark effect is directly observed in the spectral domain and in the temporal one. Varying the pump intensity, induced absorption, and gain alternate. The spectral shape of the sidebands and the contribution of the many-body interactions are discussed in the framework of the semiconductor Bloch equations.

The sample, grown by molecular-beam epitaxy, consists of a $\lambda/2$ GaAs cap layer, a single $In_{0.04}Ga_{0.96}As$ quantum well, a $\lambda/2$ GaAs spacer, and a 30 pairs AlAs/AlGaAs distributed Bragg reflector before the GaAs substrate (λ is the wavelength corresponding to the excitonic transition). The Bragg mirror makes the incident and reflected laser beams constructively interfere at the quantum-well position, thus enhancing the effective electric field. This arrangement gives an excitonic linear absorption peak as high as 70%, which is much stronger than the typical values of single GaAs-based quantum wells ($\simeq 2\%$). The transmitted intensity is small compared to the reflected one (less than 1% of the incident intensity) and the Bragg mirror stop band is almost flat in the

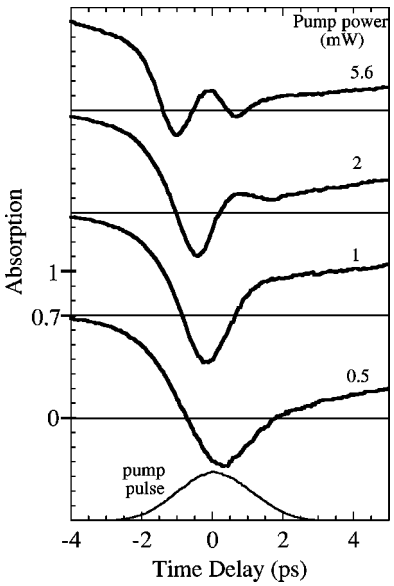

FIG. 1. Absorption at the exciton energy as a function of the pump to probe delay, for growing pump powers. The signal is detected in a 0.7 meV-wide spectral window. For each intensity one thin line marks the unperturbed absorption value ($A \simeq 0.7$), one marks the zero absorption: above this line the light is absorbed, below is amplified (gain). The zero absorption line for one intensity curve is the unperturbed value for the next higher intensity curve.

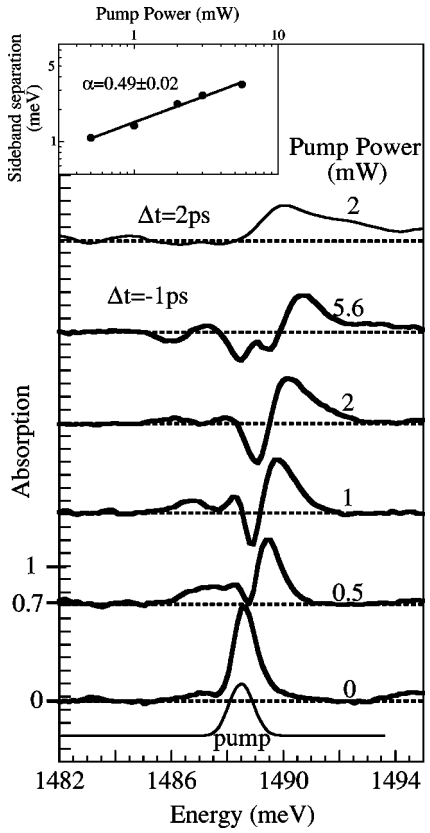

FIG. 2. Absorption spectra for growing pump powers at a fixed delay of -1 ps. On the top is shown a typical spectrum at a positive delay. Inset: spectral separation between the sidebands maxima as a function of the pump power. A least-squares fit to the data with a power dependence (solid line, α is the fitted power) indicates a square-root dependence.

spectral range we investigated; thus the absorption A can be accurately calculated from the reflectivity R: $A \simeq 1-R$. Transfer-matrix calculations confirmed that the sample structure enhances the light-exciton interaction, but does not modify it qualitatively: the excitonic absorption has a single peak and no cavity quasiparticles are created. The price to pay for the enhanced absorption is a fast re-emission of the light outside the sample, and therefore a fast radiative dephasing and line broadening [1 meV full width at half maximum (FWHM) in the linear regime, Fig. 2].

The measurements are taken in the reflection pump-probe geometry: the pump beam hits the sample at normal incidence, the probe direction makes instead a small angle with the pump ($\simeq 2°$) in order to easily discriminate the reflected beam. The intense 100 fs-long pump pulses, provided by a Ti:Sapphire laser, are spectrally filtered with a pulse shaper in order to match the exciton line (1 meV FWHM, 2 ps-long pulses) and to not overlap the absorption continuum (see Fig. 2). Given the estimate laser spot diameter of 50 μm, 1 mW of average laser power approximately corresponds to 2×10^{12} phs$\times$cm^{-2} pulse^{-1}. The excited spot in the sample is magnified and imaged on a pin hole in order to select the central region, which is almost homogeneously excited. A broadband weak pulse ($\simeq 100$ fs pulse duration, 20 meV spectral width FWHM) from the same laser tests the sample.

The absorption at the exciton energy is reported in Fig. 1 as a function of the pump to probe delay. Already for a 0.5 mW pump power, the pump pulse bleaches the excitonic absorption and induces an optical gain. This coherent gain is comparable in intensity to the linear absorption and disappears with the pump pulse. By increasing the excitation power up to 2 mW the Rabi oscillations around zero absorption are clearly resolved. The amplitude of the oscillations is maximum for small negative pump to probe delays. For positive delays the oscillations undergo a fast damping and only a long-decaying reduction of the absorption is left, due to the incoherent density effect on the excitonic transition.

In order to detect the strongest effect of the pump pulse on the absorption spectrum we fixed the delay time to -1 ps, corresponding to the maximum of the Rabi oscillation obtained for the highest pump power. The spectra are shown in Fig. 2. Initially a spectral hole is burnt at the pump peak energy and the exciton line is split into two sidebands. Then the two sidebands, comparable in intensity to the linear absorption peak, become dispersive and separate more and more for growing pump powers. The separation between the sidebands scales linearly with the pump electric field (inset Fig. 2), a typical feature of the ac Stark effect. The absorption in the spectral range excited by the pump starts being positive, then there is gain for intermediate excitations and for the highest one the gain is almost completely bleached. The high-energy sideband is much more intense than the low-energy one, so that the spectrum is not symmetric as in atomic systems. The visibility of the Stark effect is maximum when the probe pulse arrives 1 ps before the pump pulse center (-1 ps delay time). When the delay approaches zero the incoherent density effects become important: the dephasing induced by the density of excitons (or free carriers) smoothes and broadens the spectral features

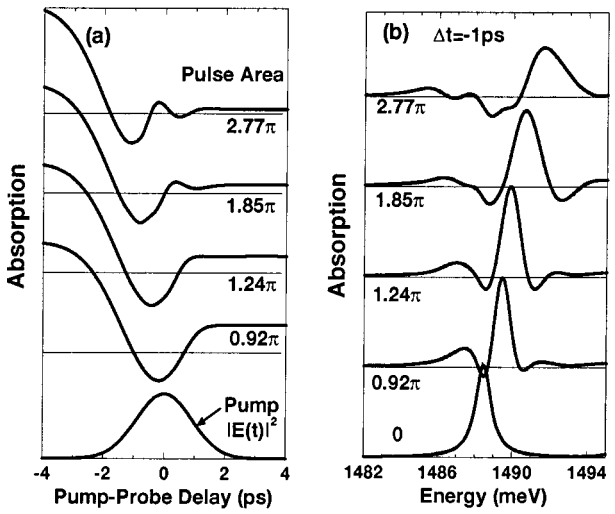

FIG. 3. (a) Calculated absorption at the pump peak frequency as a function of the pump-probe delay, for growing pump pulse areas (spectral bandpass 0.8 meV). The pump pulse is 2 ps long. The straight lines indicate zero absorption. (b) Corresponding absorption spectra showing the ac Stark splitting.

and the splitting is no longer resolved.[11] The effect is fully coherent, as the spectral modulations very rapidly disappear when the pump pulse is over: 2 ps after the pump peak only a broadening is left and an overall blueshift of the exciton line, due to the persistence of an incoherent density of excited population (see the spectrum in Fig. 2). For larger negative delays instead, the pump field is not intense enough to induce the nonperturbative effect. The highest pump intensity in our experiment is limited by the output power of the laser.

The excitation-induced dephasing grows with the excited population density, but its magnitude has an upper limit, due to the optical saturation of the transition. There is therefore an excitation regime where raising the pump field does not increase the dephasing rate, but only the Rabi frequency. In order to describe this regime, we adopted the semiconductor Bloch equations framework, which fully accounts for the coherent light-matter coupling, and we solved the equations at the Hartree-Fock approximation level, where the Coulomb interaction is included in the mean field and the higher-order many-body correlation is neglected.[17] The real sample is modeled by an effective single quantum well, without propagation effects. The exciton binding energy is taken to be $\simeq 6$ meV and the excitonic linear absorption peak linewidth (FWHM) is set to 1 meV, as the measured one, by adjusting the dephasing time in the Bloch equations. For the nonlinear regime (pump beam on) the dephasing rate is assumed to double with respect to the linear regime (2 meV). This rate is kept constant for all pump intensities, as in the saturation regime the excited population (and therefore the induced dephasing) does not significantly increase with the exciting power. A Gaussian shape is taken for the 2 ps-long pump pulse.

In Fig. 3(a) is displayed the absorption at the exciton energy as a function of the pump to probe delay, to be compared to the measured data in Fig. 1. The Rabi oscillations are clearly observable in the calculation results and the dynamics of the experimental data is reproduced. The pulse area needed to achieve the same fastest measured oscillation period (1.75 ps in Fig. 1, obtained with 5.6 mW pump power) is $4/\hbar \int \mu E dt = 2.77\pi$ (μ is the dipole moment of the electronic system, E is the electric field, t is time).[18] With this definition, in the absence of any interaction between excited particles, a 2π pulse induces one complete Rabi oscillation. But, due to the interaction, the field experienced by the electronic system is the sum of the external one and the local field generated by the coherently excited interband polarization. This contribution is of the same order as the external field, as the final result of a 2π pulse is to generate approximately two complete oscillations. Because of the dephasing, the number of observed oscillations depends not only on the pulse area but also on the pulse duration. Keeping constant the pulse area, the calculations (not shown) indicate that with a longer pulse more (but smoother) oscillations can be observed before the coherence is lost. In other words, the damping is more efficient for shorter pulses.

A series of spectra is calculated for growing pump pulse areas when the probe pulses are delayed by -1 ps with respect to the pump ones. This delay corresponds to the position of the first maximum of the Rabi oscillations in the highest pump intensity case [Fig. 3(a), pulse area 2.77π], as in the experiment. The pulse area are scaled approximately as the experimental pump fields (the area is proportional to the square root of the power). All the main features observed in the experimental spectra are reproduced by the calculations, including the coherent gain and the asymmetric amplitude of the two sidebands. In a two-level atomic system the two ac Stark sidebands have the same amplitude, while in the semiconductor the high-energy sideband is observed to be much more intense than the low-energy one. This asymmetry can be attributed to the presence of the continuum states nearly resonant with the high-energy sideband. Under intense optical pumping of the exciton transition, the continuum states tend to redshift, coming closer in energy to the high-energy sideband and enhancing it. Another contribution can come from the exciton blueshift, which occurs in a dense gas because of the repulsive exciton-exciton interaction. Furthermore, the Hartree-Fock approximation is known to overestimate the importance of the continuum states, as the excitonic correlation is only partially included. The enhancement of the high-energy sideband is indeed even more pronounced in the calculated spectra than in the experimental ones.

The measured and calculated spectral sidebands are reminiscent of the Stark effect of a two-level transition excited by a cw laser.[1,3] But in our conditions the electric field strongly varies in time and the spectrum is distorted by the peculiar features of the semiconductor, so that we could not associate to each spectrum a precise Rabi energy. Furthermore the pump pulse duration is comparable to the Rabi period, so that only a mean Rabi energy can be inferred from the period of the oscillations in Figs. 1 and 3(a). The half spectral separations between the two sidebands, as well as the inverse period of the oscillations in the delay time, give anyway the order of magnitude of the Rabi energy generating the effect.

In conclusion we report the direct observation of the optical Stark splitting in the excitonic absorption spectrum of a single semiconductor quantum well. The ac Stark sidebands

are as large as half of the linear absorption peak and disappear in less than 2 ps. The semiconductor Bloch equations in the Hartree-Fock approximation show that the effect is qualitatively due to the coherent exciton-light coupling and the Coulomb dephasing just provides small corrections.

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[18] In the semiconductor community the pulse area is sometimes defined as $2/\hbar \int \mu E dt$, which is two times smaller than the quantity we borrowed from atomic physics.