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Dressed excitons within an incoherent electron gas: Observation of a Mollow triplet and an Autler-Townes doublet

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We demonstrate that the interaction between excitons and a sea of incoherent electrons does not preclude excitons dressing by light. We investigate if exciton-electron scattering plays some inhibiting role in the coherent light-exciton coupling by measuring the dynamical absorption spectrum of a modulation-doped CdTe quantum well that shows clear evidence for significant electron scattering of the excitonic states. We show the occurrence of dressed and correlated excitons by detecting quantum coherent interferences through excitonic Autler-Townes doublet and ac Stark splitting that evolves into a Mollow triplet with gain. We also evidence the partial inhibition of the electron-exciton scattering by exciton-light coupling.

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The absorption spectrum of semiconductor quantum wells is dominated by a strong exciton resonance. Excitons are bound, conduction electron valence-hole pairs and are quite often compared to a two level system as a hydrogen atom. The role of many body interactions and exciton correlations in the exciton dynamical absorption spectrum has attracted much interest for several decades. The response of the exciton absorption to an incoherent and thermalized electron-hole population obtained through nonresonant pumping in the continuum corresponds mainly to a broadening of the exciton resonance that is dominated by collision broadening (excitation-induced dephasing) rather than by a bleaching due to the saturation of the oscillator strength caused by phase space filling.[1] The effect increases with electron-hole concentration. In the regime of coherent pumping at the exciton resonance, the light-field dresses the electron-hole gas and dynamical Stark splitting has been clearly observed in GaAs quantum wells embedded[2] or not[3,4] in a microcavity. Actually, in this coherent regime, the excitation-induced dephasing is found to be reduced as a result of the significant decrease of the Coulomb collision rates.[5] As a consequence, the light-induced exciton dressing renders this many-body excitonic system comparable to a noninteracting atom system.[6] This issue is very important if one tries to extend coherent manipulations as those performed in atomic systems[7] to semiconductors in view of possible applications.

In modulation-doped semiconductor quantum wells, excess carriers coalesce with excitons to form charged excitons (trions). The absorption spectrum is then modified: a trion resonance appears below the exciton resonance.[8] However, many-body interactions also tend to become more complex. Electrons, for instance, are known to strongly screen the exciton oscillator strength[8,9] and to modify their linewidth and binding energy through exchange interactions.[10] Thus, the shape of the exciton line, as well as that of the trion line, depends strongly on the strength of electron scattering with excitons and trions. Indeed, we have observed, as others before us, that a high-energy tail develops at both exciton and trion resonances upon increasing electron concentration.[11] This gives strong evidence for the strength of the interaction of electrons with excitons and trions. In this condition, it is expected that coherent exciton-light coupling should be destroyed due to electron-exciton collision broadening, and profoundly change the importance of light dressing on excitons. Actually, we evidence that the nonlinear exciton-light coupling dominates the electron-exciton scattering. This fact brings new openings towards possible coherent manipulations in semiconductor quantum wells containing an electron gas.

In this paper, we investigate the exciton light dressing in a sea of thermalized electrons. We selected a sample which evidences electron scattering with excitons and trions through a prominent high-energy tail in their absorption resonances. We use a pump pulse to coherently prepare the exciton state and we follow the evolution of the excitonic absorption line by measuring the spectrum of a weak probe pulse at different delays with respect to the preparing pump pulse. Detecting the excitonic ac Stark splitting, Mollow triplet and Autler-Townes doublet allows us to demonstrate that the interaction between thermalized electrons and excitons does not prevent dressing excitons. In addition, we evidence that exciton-light coupling partially inhibits scattering of excitons by electrons. This is established by exciton resonance narrowing and reduction of the high-energy absorption tail.

Possible exciton transitions are determined by the optical selection rules. In Fig. 1(a), we show the diagram for exciton-light coupling for pulsed σ^+ and σ^- circular-polarizations, considering both excitons (X) and biexcitons bound (XX_b) and unbound (XX_u) states. The heavy-hole exciton state X_{σ^+} is dressed by a σ^+ pump pulse and probed by either a σ^+ [Fig. 1(b)] or a σ^- [Fig. 1(c)] probe pulse. Figure 1(d) shows the scheme for dressing the bound-biexciton state XX_b with a σ^+ pump pulse and probing the exciton state X_{σ^-}.

We used a 8 nm modulation doped CdTe quantum well (4×10^{10} cm^{-2}),[11,12] with an exciton line at 1625.7 meV and a trion line at 1622.4 meV. The high-energy tail of both exciton and trion resonances is broadenned by electron-exciton scattering (see the bottom panel of Fig. 2) which can be reproduced adequately by our calculations (see Ref. 11 for details). Our calculations further indicate that exchange scattering is stronger than direct Coulomb scattering (see also, Refs. 10 and 13), and therefore dominates the broadening mechanisms. At 5 K, pump and probe experiments were performed with a narrow tuneable 4 ps pump and a broad 100 fs

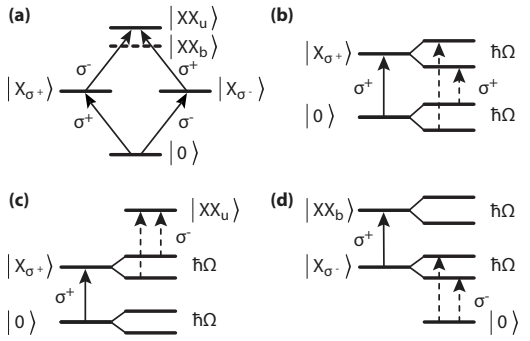

FIG. 1. Exciton-light coupling diagram, respecting exciton-transition selection rules (a) σ^+ (σ^-) spin-up +1 (spin-down −1) exciton state X_{σ^+} (X_{σ^-}) with $-1/2$ ($+1/2$) electron spin and hole angular momentum projection $+3/2$ ($-3/2$). (b) σ^+ pump pulse dresses X_{σ^+} state which is probed by a σ^+ probe pulse, when it is probed by a σ^- pulse, the transition is to unbound XX_u state (c). (d) The bound XX_b is dressed by a σ^+ pump pulse and the X_{σ^-} state is probed by a σ^- pulse. $\hbar\Omega$ represents the ac Rabi splitting of the dressed eigenstates.

probe with an intensity ratio larger than 50. We recorded the reflectivity (Fig. 2) and differential reflectivity (Figs. 3 and 4) for various delays between pump and probe pulses.

In our investigations, we use the pump pulse to dress exciton states and we are interested to follow their evolution in time. It is important to know that dressed states are best evidenced when the probe and pump pulses overlap at negative time delays.[2,3] Then, we use pump pulses with 4 ps duration and with the highest possible intensity (about 2×10^{12} photons/pulse cm^2).

First, we tuned the pump pulse on the exciton resonance and we worked in the $\sigma^+\sigma^+$ polarization configuration [Fig. 1(b)]. As the probe delay is changed at negative delays, we monitored the build up of the pump field and its effect on the exciton resonance. As Fig. 2(a) shows, the exciton resonance splits up as the pump intensity rises, i.e., as the delay time goes to zero. Two side modes grow apart, with an energy

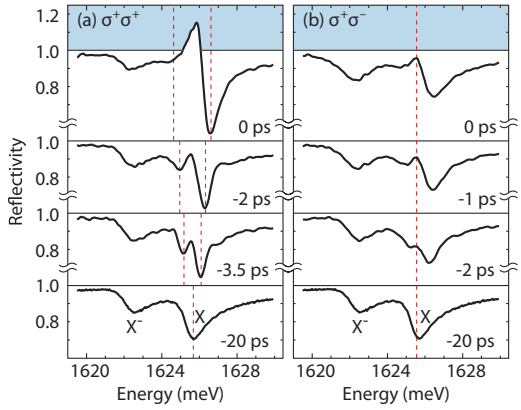

FIG. 2. (Color online) Reflectivity spectra collected at various delay times for a σ^+ pump tuned to the exciton resonance (1625.7 meV). The vertical scale is the same for all spectra. (a) σ^+ probe, the dashed lines indicate the position of the Stark split modes. (b) σ^+ probe, the dashed-line indicates the position of the pump pulse.

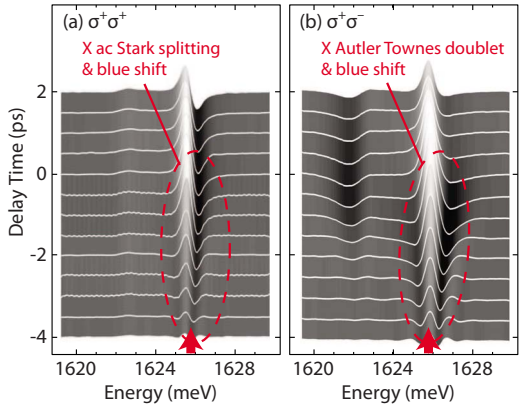

FIG. 3. (Color online) Differential reflectivity spectra as a function of the delay time for (a) $\sigma^+\sigma^+$ and (b) $\sigma^+\sigma^-$ pump and probe. The σ^+ pump (vertical arrow) is tuned to the exciton resonance (1625.7 meV).

separation scaling with the square root of the pump intensity. This is a key feature of the ac Stark splitting effect, which originates from a nonlinear coupling between the strong electromagnetic field and the excitonic resonance. This coherent light-exciton coupling is characterized by a Rabi energy $\hbar\Omega$. The Rabi energy is half of the separation between the sidebands which correspond to transitions between the eigenstates of the exciton system dressed by the electromagnetic field [Fig. 1(b)]. Would the exciton-electron scattering be very efficient, the exciton dephasing time would be very fast thereby destroying the coherent exciton-light coupling. In such a condition, the ac Stark splitting would not be observable. The clear observation of the dressed-exciton in the optical response of a quantum well containing free carriers indicates that the nonlinear exciton-light coupling dominates the electron-exciton scattering. Upon excitation we clearly observe a narrowing of the Stark sidebands together with a reduction of the high-energy tail. It is important to remind that this high-energy absorption is due to exchange electron-exciton scattering. According to this discussion, our result

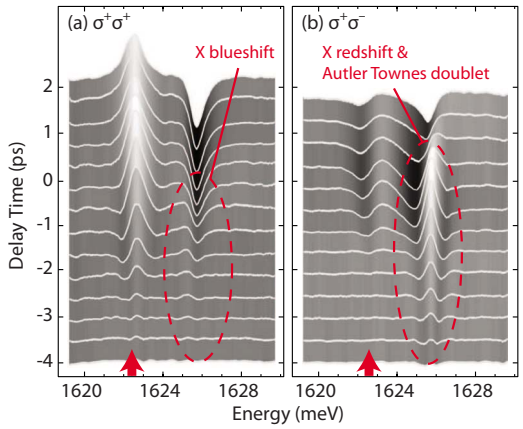

FIG. 4. (Color online) Differential reflectivity spectra as a function of the delay time for (a) $\sigma^+\sigma^+$ and (b) $\sigma^+\sigma^-$ pump and probe. The σ^+ pump (vertical arrow) is tuned close to the biexciton resonance (1621.8 meV).

evidences that the dressing of excitons by light dramatically reduces their exchange interaction with the thermalized electrons.

Interestingly enough, the original exciton spectral position becomes increasingly transparent as the pump power increases and eventually the transparency turns into gain as in a strongly driven two-level system. Mollow predicted this gain without inversion of population;[14] it can be seen as interferences of various paths of n photons transitions between dressed states. Ciuti *et al.*[5,15] have shown that this simple two-level picture still holds for the much more complex case of excitons in a quantum well, allowing for the existence of gain. This gain observed here evidences the occurrence of quantum coherent interferences via dressed exciton states, even in the presence of thermalized electrons.

Of course, the pump field is also absorbed, which creates excitons. Excitons repel each other due to Pauli exclusion principle through exchange of their constituent carriers. The exciton resonance blueshifts,[16] rendering the exciton Stark split states asymmetric, as we observe. The build up of the exciton blue shift is clearer on differential reflectivity spectra [Fig. 3(a)]. Similar to ac Stark splitting, the role of electrons on exciton-exciton correlations of same spin through exchange interaction is not preponderant. Still, their signature is very clear at positive delay: they coalesce with excitons to form trions within 10 ps.[17]

Since the σ^+ pump field only couples to exciton with angular momentum +1, ac Stark splitting should not be observed for $\sigma^+\sigma^-$ counter circular polarizations because the σ^+ and σ^- transitions share no common states [Fig. 1(a)]. However, if excitons with opposite spins are correlated, they can form bound and unbound biexciton states [see three-level system in Fig. 1(a)]. Then, although σ^+ and σ^- transitions share no common state, the σ^+ pump can dress the X_{σ^+} state and the probe may coherently couple this X_{σ^+} dressed state to the unbound biexciton state XX_u [Fig. 1(c)]. Indeed, as the pump field builds up, a spectral dip appears in the probe reflectivity spectrum at the exciton energy [Fig. 2(b)]. At zero delay, the sample even becomes fully transparent at this wavelength. So the probe reflectivity spectrum features two resonances called Autler-Townes doublet.[18] This doublet may be traced back to both transitions from X_{σ^+} dressed state to undressed unbound biexciton state XX_u [Fig. 1(c)]; the Rabi energy being the separation between the resonances, which is half the energy separation between the sidebands in ac Stark splitting. Indeed, we find a smaller energy separation between the two sidebands for countercircular polarization than for co-circular polarization. However, the measured energy is not exactly the expected value, which is not surprising considering that this three-level system is formed by correlated exciton and biexciton states together with their renormalization energies. This result would require a full theoretical analysis, which is not the scope of this paper.

Due to the fact that Autler-Townes doublet is detected by probing the transitions from eigenstates of the dressed exciton to undressed biexciton state, it should be much more sensitive to the presence of electrons than in the case of ac Stark splitting. Actually, this is revealed by the tiny decrease of the high energy tail of their sidebands.

More insight on the exciton correlations is gained by looking at the differential spectra in Fig. 3(b). The observed nonlinearities on the exciton resonance at short positive delay times are very much comparable to the ones observed for $\sigma^+\sigma^+$ pump and probe [Fig. 3(a)]; a blueshift of the exciton resonance as well as a strong decrease of the exciton resonance is clearly visible. Yet, neither Pauli blocking nor first-order Coulomb-induced nonlinearity are expected to lead to a coupling among the subspaces of different exciton spin state, thus, the observed correlations evidence high-order Coulomb correlations between excitons.[19] Their strength makes the observation of biexciton bound states likely[20] and we associate the induced absorption that shows up about 4 meV below the exciton line to bound biexcitons.[21]

Since bound biexciton states can be formed within an electron gas, we can now determine if, similarly to unbound biexciton states, they may take part into coherent processes. To this aim, we tune the pump close to the bound-biexciton resonance in $\sigma^+\sigma^-$ configuration.[28] At exciton resonance and negative delay times, differential spectra [Fig. 4(b)] evidence a coherent signal similar to the one observed in Fig. 3(b). Despite the electron population, the virtual bound-biexciton state XX_b is dressed by the σ^+ pump pulse and the Autler-Townes doublet can be probed via the X_{σ^-} exciton transition [Fig. 1(d)]. This is possible because the X_{σ^-} exciton and the σ^+ dressed XX_b biexciton share a common state. In the case of $\sigma^+\sigma^+$ configuration, bound biexcitons cannot be formed and no coherent signal is established at exciton resonance [Fig. 4(a)].

This result is corroborated by the observation, close to zero delay, of the excitonic optical Stark shift,[22–25] which is due to a coupling between the exciton and all virtual two-exciton states, bound and unbound. At small detuning, when the pump is tuned close to biexciton energy, this coupling is mainly due to Coulomb interaction. It leads to a redshift of the exciton line in the case of stable bound biexciton state,[25] otherwise the exciton resonance undergoes a blueshift. Indeed, the exciton resonance is blueshifted in $\sigma^+\sigma^+$ configuration [Fig. 4(a)], while in $\sigma^+\sigma^-$ it is redshifted [Fig. 4(b)]. When the pump is tuned close to the biexciton resonance, it overlaps inevitably with the trion resonance and a population of trions is generated, depleting the well from its excess electrons. This diminishes the scattering of excitons by electrons. As a consequence, both excitonic blueshift and redshift evolve into a net excitonic induced absorption. In addition, a reduced absorption of the excitonic high-energy tail is observed for $\sigma^+\sigma^-$ configuration evidencing the reduction of the exciton-electron exchange scattering.[11,26]

Note that by applying a light field resonantly to the exciton resonance, in a doped semiconductor quantum well, the transition can be coupled to the optical external field as well as it can experience internal interactions with the electrons. When the applied light field is weak, the internal field dominates and we measure the linear response to the weak perturbation, which is the excitonic absorption broadening mainly due to exciton-electron exchange interaction. It is important to comment that the intensity needed in undoped semiconductor quantum wells[3,27] is in the same order of magnitude as that used in this work to get an equivalent ac Stark effect. This means that the nonlinear light-exciton coupling dominates over the internal exciton-electron interaction. This re-

veals that exciton-electron interactions have no inhibiting role in the build-up of the coherence effects. This probably comes from the mixed electronic and photonic nature of the dressed state: its partially photonic (bosonic) nature inefficiently scatters with electrons. This inefficiency of scattering we have also evidenced by the narrowing of the Stark sidebands.

As a conclusion, we gave evidence in this work that coupling of excitons to the light field dominates over the exchange scattering between excitons and electrons even in a sea of thermalized electrons. We have observed, in a modulation doped quantum well, a clear ac Stark splitting, which evolves in a Mollow triplet with gain at exciton resonance. In addition, we demonstrated that exciton correlations remain very strong within an electron gas. They yield unbound and stable bound biexciton pairs that play an important role in quantum coherent processes evidenced via Autler-Townes doublet and optical Stark shift. We also showed a narrowing of the exciton sidebands, evidencing the lowering of scattering between incoherent electrons and dressed excitons.

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[1] F. Jahnke *et al.*, Phys. Rev. Lett. **77**, 5257 (1996).
[2] F. Quochi, G. Bongiovanni, A. Mura, J. L. Staehli, B. Deveaud, R. P. Stanley, U. Oesterle, and R. Houdré, Phys. Rev. Lett. **80**, 4733 (1998).
[3] M. Saba, F. Quochi, C. Ciuti, D. Martin, J.-L. Staehli, B. Deveaud, A. Mura, and G. Bongiovanni, Phys. Rev. B **62**, R16322 (2000).
[4] A. Schülzgen, R. Binder, M. E. Donovan, M. Lindberg, K. Wundke, H. M. Gibbs, G. Khitrova, and N. Peyghambarian, Phys. Rev. Lett. **82**, 2346 (1999).
[5] C. Ciuti, C. Piermarocchi, V. Savona, P. E. Selbmann, P. Schwendimann, and A. Quattropani, Phys. Rev. Lett. **84**, 1752 (2000).
[6] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990), Vol. 1.
[7] M. Fleischhauer, A. Imamoglu, and J. P. Marangos, Rev. Mod. Phys. **77**, 633 (2005).
[8] K. Kheng, R. T. Cox, Y. Merle d'Aubigné, F. Bassani, K. Saminadayar, and S. Tatarenko, Phys. Rev. Lett. **71**, 1752 (1993).
[9] V. Huard, R. T. Cox, K. Saminadayar, A. Arnoult, and S. Tatarenko, Phys. Rev. Lett. **84**, 187 (2000).
[10] G. Ramon, A. Mann, and E. Cohen, Phys. Rev. B **67**, 045323 (2003).
[11] J. Berney, L. Schifferle, M. T. Portella-Oberli, and B. Deveaud, e-print arXiv:0712.1248.
[12] T. Wojtowicz, M. Kutrowski, G. Karczewski, and J. Kossut, Appl. Phys. Lett. **73**, 1379 (1998).
[13] C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **58**, 7926 (1998).
[14] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).
[15] C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **58**, 7926 (1998).
[16] S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).
[17] M. T. Portella-Oberli, V. Ciulin, M. Kutrowski, T. Wojtowicz, and B. Deveaud, Phys. Status Solidi B **238**, 513 (2003).
[18] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).
[19] T. Meier, S. W. Koch, P. Brick, C. Ell, G. Khitrova, and H. M. Gibbs, Phys. Rev. B **62**, 4218 (2000).
[20] U. Neukirch, S. R. Bolton, L. J. Sham, and D. S. Chemla, Phys. Rev. B **61**, R7835 (2000).
[21] P. Kossacki, P. Plochocka, B. Piechal, W. Maslana, A. Golnik, J. Cibert, S. Tatarenko, and J. A. Gaj, Phys. Rev. B **72**, 035340 (2005).
[22] A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoc, Phys. Rev. Lett. **56**, 2748 (1986).
[23] D. Hulin and M. Joffre, Phys. Rev. Lett. **65**, 3425 (1990).
[24] M. Combescot and R. Combescot, Phys. Rev. Lett. **61**, 117 (1988).
[25] M. Combescot and R. Combescot, Phys. Rev. B **40**, 3788 (1989).
[26] M. T. Portella-Oberli, J. Berney, B. Deveaud, V. Ciulin, M. Kutrowski, and T. Wojtowicz, Acta Phys. Pol. A **106**, 115337 (2004).
[27] M. C. Phillips and H. Wang, Phys. Rev. B **69**, 115337 (2004).
[28] When off resonant, nonlinearities are small and it is more convenient to assess them through the study of differential reflectivity spectra.