Strongly Driven Exciton Resonances in Quantum Wells: Light-Induced Dressing versus Coulomb Scattering

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The nonequilibrium dynamics of a two-dimensional electron-hole gas is studied in the regime of strong and resonant pumping of the exciton resonance. The Coulomb collision rates are consistently determined by taking into account the light-induced coherence of the two-band system that leads to a dressing of the carrier spectral functions. The light dressing dramatically reduces the Coulomb scattering efficiency. Results are presented for Rabi oscillations in the time domain and dynamical Stark splitting in the pump-probe absorption spectra.

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The optical coherent effects due to the nonlinear light-matter interaction are well established in atomic and molecular systems [1]. In particular, when a strong resonant pulse coherently saturates an atomic transition, transient Rabi oscillations of the excited population occur in the time domain. Spectrally, the absorption of a weak probe beam shows coherent sidebands, known as dynamical Stark splitting. These coherent features can be dramatically frustrated by relaxation and dephasing. However, in a very low pressure atomic gas, a nearly collisionless regime can be achieved leading to the observation of spectacular coherent saturation effects [2].

In solid state physics, the situation is much more intriguing [3,4]. In fact, in a semiconductor system the density of photon-generated electron-hole pairs increases with the pump intensity as well as the Coulomb collision rates (excitation-induced dephasing). In a recent Letter, Jahnke *et al.* [5] studied the bleaching of the exciton resonance in the presence of an *incoherent* and *thermalized* electron-hole plasma. Such a situation can be obtained through nonresonant pumping in the continuum absorption. Both theory and experiment show that, in such an incoherent regime, the bleaching of the exciton absorption is dominated by the collision broadening rather than by the saturation of the oscillator strength (phase space filling).

However, more recent experiments [6–8] have shown a very different role for the excitation-induced dephasing in the regime of *coherent pumping* at the exciton resonance. In particular, Quochi *et al.* [6] reported the first observation of the dynamical Stark splitting in semiconductors. Several well-resolved Rabi oscillations have also been observed in the time domain. These coherent effects have been observed in GaAs quantum wells embedded in a microcavity. Indeed, the spectral filter provided by the cavity is crucial in the exciton saturation regime. In fact, it allows one to excite resonantly the exciton transition, limiting the (undesired) creation of electron-hole pairs with large excess kinetic energies. In an even more recent paper, Schülzgen *et al.* [8] reported the observation of several Rabi oscillations in free-space GaAs quantum wells, using spectrally narrow pump pulses whose width ($\sim$1 meV) is comparable to the cavity linewidth in Refs. [6,9].

In the above-mentioned experiments [6–8], the Rabi energy of the driving electromagnetic field is comparable to the exciton binding energy. This simply means that the light-field dressing of the electron-hole gas can not be neglected. As shown by Schmitt-Rink *et al.* [10], the interband coherence causes a drastic modification of the carrier spectral functions. Different carrier spectral functions lead to different collision rates. In the case of interaction with optical phonons, Tran Thoai and Haug [11] have included the light dressing in their calculations. They have actually found that LO-phonon quantum beats are much more pronounced when the dressed spectral functions are consistently included. In the case of strongly driven resonances at very low temperatures, the scattering mechanism is provided by Coulomb interaction.

In this Letter, we study the role of Coulomb scattering in the coherent transients of strongly driven exciton resonances. We derive the dressed semiconductor Bloch equations for carrier populations and interband polarization. Within a nonequilibrium formalism [4,12,13], Coulomb scattering is treated in the second Born approximation and the light dressing of the carrier spectral functions is consistently included. The light dressing is found to significantly decrease the Coulomb collision rates. For coherent excitation at the exciton resonance, Coulomb scattering is not strong enough to quench the coherent saturation effects due to phase space filling. In particular, our calculation show a dynamical Stark splitting in the absorption spectrum of a weak probe beam.

The physical quantity which we are going to study is the two-by-two density matrix:

$$\rho_{\mathbf{k}}(t) = \begin{pmatrix} n_{1,\mathbf{k}}(t) & P_{\mathbf{k}}(t) \\ P^{\star}_{\mathbf{k}}(t) & n_{2,\mathbf{k}}(t) \end{pmatrix}.$$

The diagonal elements $n_{1,\mathbf{k}}(t)$, $n_{2,\mathbf{k}}(t)$ are the occupation

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numbers of the conduction and valence $\mathbf{k}$ states, respectively. The off-diagonal term $P_{\mathbf{k}}(t)$ is the interband polarization. We do omit the spin index because we consider an exciting field with a definite circular polarization and neglect spin relaxation. The equation of motion for the density matrix is the sum of two terms:

$$\frac{\partial \rho_{\mathbf{k}}(t)}{\partial t} = \frac{\partial \rho_{\mathbf{k}}(t)}{\partial t}\bigg|_{\text{coher}} + \frac{\partial \rho_{\mathbf{k}}(t)}{\partial t}\bigg|_{\text{scatt}}.$$

The first term gives rise to the Hartree-Fock semiconductor Bloch equations [10]. In other words, it contains the coupling with the external (classical) light field and the first-order Coulomb contribution (Hartree-Fock). The Hartree-Fock equations are fully coherent, and atomiclike saturation effects take place due to the phase space filling nonlinearity, namely, field-driven Rabi oscillations [14] and dynamical Stark splitting [15]. The second term represents the Coulomb collision rate. To calculate this term, one has to deal with the spectral functions or, more precisely, with the retarded Green's function $G^r_{\mathbf{k}}(t,t')$ which is a two-by-two matrix in a two-band system. Usually, a *diagonal* approximation for $G^r_{\mathbf{k}}(t,t')$ is introduced [5,16]:

$$G^r_{\mathbf{k}}(t,t') = -i\theta(t-t')\left\{\begin{pmatrix}1 & 0\\ 0 & 0\end{pmatrix} e^{-i(\epsilon_{1,\mathbf{k}}-i0^+)(t-t')} + \begin{pmatrix}0 & 0\\ 0 & 1\end{pmatrix} e^{-i(\epsilon_{2,\mathbf{k}}-i0^+)(t-t')}\right\}. \tag{1}$$

The quantities $\hbar\epsilon_{1,\mathbf{k}}(T)$ and $\hbar\epsilon_{2,\mathbf{k}}(T)$ are, respectively, the conduction and valence band energy dispersions with the Hartree-Fock intraband renormalization. Explicitly, we have ($j = 1,2$):

$$\epsilon_{j,\mathbf{k}}(T) = \frac{\omega_g - \omega_p}{2(-1)^{j+1}} + \frac{\hbar k^2}{2m_j} - \sum_{\mathbf{q}} V_q[n_{j,\mathbf{k}-\mathbf{q}}(T) - \delta_{j,2}],$$

with $m_1 = m_e$, $m_2 = -m_h$. The quantity ω_p is the rotating frame frequency and is chosen to coincide with the central frequency of the pump pulse, while $\hbar\omega_g$ is the band-gap energy in the linear regime (unexcited crystal) and $\hbar V_q$ is the Fourier transform of the bare Coulomb potential. Notice that neither the interband polarization P_k nor the electric field $E(t)$ appear in Eq. (1). Such a spectral function is *undressed* because the light-induced renormalizations are neglected. This approximation is justified only in the case of an incoherent electron-hole population. In the coherent regime, the light-induced renormalizations have to be consistently included. Within the rotating frame approximation, the solution of the Dyson equation [10] for $G^r_{\mathbf{k}}(t,t')$, including the self-energy of the light-field and the Hartree-Fock contribution is

$$G^r_{\mathbf{k}}(t,t') = -i\theta(t-t') \sum_{A\in\{+,-\}} g_{A,\mathbf{k}}(T) e^{-i(\epsilon_{A,\mathbf{k}}-i0^+)(t-t')}, \tag{2}$$

where T is the center time $(t+t')/2$. The matrices $g_{\pm,\mathbf{k}}(T)$ are defined as

$$g_{\pm,\mathbf{k}}(T) = \frac{1}{\Delta_{\mathbf{k}}(T)} \times \begin{pmatrix} \epsilon_{\pm,\mathbf{k}}(T) - \epsilon_{2,\mathbf{k}}(T) & \tilde{\Omega}_{\mathbf{k}}(T) \\ \tilde{\Omega}^{\star}_{\mathbf{k}}(T) & \epsilon_{\pm,\mathbf{k}}(T) - \epsilon_{1,\mathbf{k}}(T)\end{pmatrix}$$

with

$$\Delta_{\mathbf{k}}(T) = \sqrt{[\epsilon_{1,\mathbf{k}}(T) - \epsilon_{2,\mathbf{k}}(T)]^2 + 4|\tilde{\Omega}_{\mathbf{k}}(T)|^2}.$$

The dispersions of the *dressed* bands are given by

$$\epsilon_{\pm,\mathbf{k}}(T) = \frac{\epsilon_{1,\mathbf{k}}(T) + \epsilon_{2,\mathbf{k}}(T)}{2} \pm \frac{1}{2}\Delta_{\mathbf{k}}(T).$$

The band dressing is given by the interband coupling due to the effective Rabi energy, including external contribution and local field correction

$$\hbar\tilde{\Omega}_{\mathbf{k}}(T) = \mu_{cv}E(T) + \sum_{\mathbf{q}} \hbar V_q P_{\mathbf{k}-\mathbf{q}}(T),$$

where $E(T)$ is the time envelope of the driving electric field. Because of the Rabi energy, the *dressed* retarded Green's function is *nondiagonal* and *coherent*. Using the generalized Kadanoff-Baym ansatz [17] and taking the Markov limit, we find the following expression for the dressed second Born collision rate:

$$\frac{\partial \rho_{\mathbf{k}}(t)}{\partial t}\bigg|_{\text{scatt}} = \{\Gamma^{\text{in}}_{\mathbf{k}}(t) - \Gamma^{\text{out}}_{\mathbf{k}}(t)\} + \text{h.c.}, \tag{3}$$

where

$$\Gamma^{\text{out}}_{\mathbf{k}}(t) = \sum_{\mathbf{q},\mathbf{q}'} \sum_{A,B,C,D\in\{+,-\}} i\left\{ \frac{|V_q|^2 g_{A,\mathbf{k}-\mathbf{q}}(1-\rho_{\mathbf{k}-\mathbf{q}})\,\mathrm{Tr}[g_{B,\mathbf{q}'}(1-\rho_{\mathbf{q}'})\rho_{\mathbf{q}'-\mathbf{q}} g_{C,\mathbf{q}'-\mathbf{q}}]\rho_{\mathbf{k}} g_{D,\mathbf{k}}}{\epsilon_{A,\mathbf{k}-\mathbf{q}} + \epsilon_{B,\mathbf{q}'} - \epsilon_{C,\mathbf{q}'-\mathbf{q}} - \epsilon_{D,\mathbf{k}} - i0^+} - \frac{V_q V_{q'} g_{A,\mathbf{k}-\mathbf{q}}(1-\rho_{\mathbf{k}-\mathbf{q}})\rho_{\mathbf{k}-\mathbf{q}-\mathbf{q}'} g_{C,\mathbf{k}-\mathbf{q}-\mathbf{q}'} g_{B,\mathbf{k}-\mathbf{q}'}(1-\rho_{\mathbf{k}-\mathbf{q}'})\rho_{\mathbf{k}} g_{D,\mathbf{k}}}{\epsilon_{A,\mathbf{k}-\mathbf{q}} + \epsilon_{B,\mathbf{k}-\mathbf{q}'} - \epsilon_{C,\mathbf{k}-\mathbf{q}-\mathbf{q}'} - \epsilon_{D,\mathbf{k}} - i0^+}\right\}.$$

The in-scattering rate $\Gamma^{\text{in}}_{\mathbf{k}}(t)$ is obtained by the out-scattering rate $\Gamma^{\text{out}}_{\mathbf{k}}(t)$ through the replacement $\rho \rightleftharpoons (1-\rho)$. In the above expressions, we have simplified the notation by omitting the time dependence. Actually, $\rho_{\mathbf{k}} \equiv \rho_{\mathbf{k}}(t)$ and $g_{A,\mathbf{k}} \equiv g_{A,\mathbf{k}}(t)$ [18]. We have analytically checked that the sum rule $\sum_{\mathbf{k}} \partial\rho_{\mathbf{k}}(t)/\partial t|_{\text{scatt}} = 0$ is satisfied. Therefore, Coulomb scattering redistributes the density matrix at different $\mathbf{k}$. This means a redistribution of the population within each band (diagonal elements) as well as a redistribution of the interband polarization (off-diagonal element).

Now, we apply the present theory by considering the effect of a strong pump beam tuned at the exciton resonance ($\hbar\omega_p = \hbar\omega_g - E_b$). In particular, we consider a Gaussian pulse whose area is $(2/\hbar)\int dt\, \mu_{cv}E(t) = 3\pi$ and whose peak Rabi energy is $\mu_{cv}E(0) = 0.4E_b$ with E_b being the two-dimensional exciton binding energy. The pulse has an energy width (HWHM) equal to $0.24E_b$ which allows a resonant excitation of the excitonic transition. In the calculations, we consider an electron-hole mass ratio $m_e/m_h \simeq 0.4$ (GaAs quantum well). In Fig. 1, we show the results for the time evolution of the total carrier density per unit area $n(t) = \sum_{\mathbf{k}} n_{1,\mathbf{k}}/A$. The dressed Born calculation (solid line) shows well-resolved field-driven Rabi oscillations of the excited density. In the dotted line, we can see the Hartree-Fock results with a (small) constant dephasing ($\hbar/T_2 = 0.07E_b$). The undressed Born result (long-dashed line) shows a much more pronounced damping of the Rabi oscillations. Indeed, the consistent inclusion of the light dressing significantly reduces the Coulomb collision rates as in the case of interaction with optical phonons [11]. In Fig. 2, we can see directly the collision rates at the time when the dressed and undressed calculations begin to diverge (see vertical arrow in Fig. 1). Indeed, the dressed rates (solid line) are drastically smaller than the undressed ones (long-dashed line) both for population and polarization scattering. Actually, the interband coherence of the carrier spectral functions leads to a destructive interference between the different scattering channels. A physical observable which is particularly sensitive to dephasing is obviously the absorption. In Fig. 3, we show the pump-probe absorption spectrum for a weak probe pulse. Such a quantity is proportional to $\frac{\delta P(\omega)}{\delta E(\omega)}$, where $\delta P(\omega)$ is the frequency-dependent interband polarization induced by the probe field $\delta E(\omega)$. In this calculation, we have considered a degenerate probe pulse which arrives at the maximum of the pump and whose spectral width is (HWHF) $2.5E_b$. As a reference, we present also the linear regime spectrum (short-dashed line), exhibiting the sharp exciton resonance followed by the steplike continuum absorption. The dressed spectrum (solid line) shows a well-resolved dynamical Stark splitting which is the spectral domain counterpart of the field-driven Rabi oscillations in the time domain. Indeed, such a spectrum is reminiscent of the Mollow absorption spectrum occurring in the two-level system and observed in atomic systems. With respect to the atomic case, the presence of the continuum states leads to an asymmetric shape in the spectrum. In particular the higher energy sideband is enhanced. On the other hand, in the undressed Born calculation (long-dashed line), the coherent sidebands are not resolved because the excitation-induced dephasing is dramatically overestimated.

Concerning the range of application of the presented theory, two points need to be discussed: screening and Markov approximation. We have performed calculations also with quasistatic RPA screening, obtaining results which are qualitatively similar to the unscreened case. In a recent letter, Banyai *et al.* [19] studied the role of dynamical screening and non-Markovian kinetics for a three-dimensional bulk system. They have found results

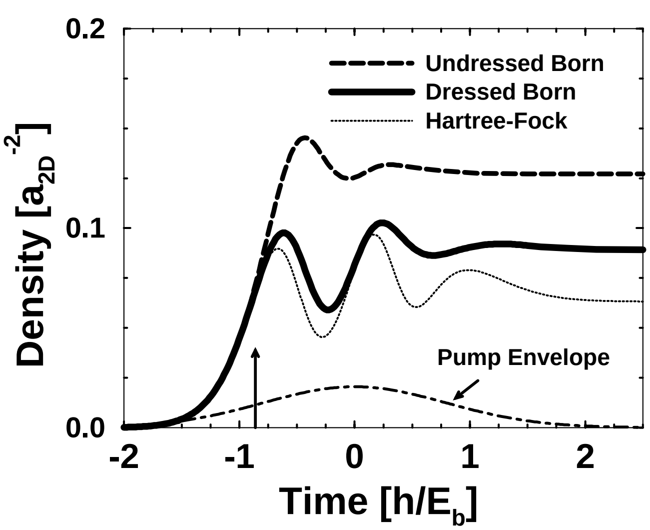

FIG. 1. Total carrier density ($1/a_{2D}^2$ units) versus time (h/E_b units). a_{2D} is the two-dimensional exciton radius ($a_{2D} = a_{3D}/2$). E_b is the 2D exciton binding energy. Dotted-dashed line: pump envelope $E(t)$ (arbitrary units). The pulse area is 3π and peak Rabi energy $\mu_{cv}E(0) = 0.4E_b$. Dotted line: Hartree-Fock with (small) constant damping ($\hbar/T_2 = 0.07E_b$). Solid line: dressed Born. Long-dashed line: undressed Born. The vertical arrow indicates the time examined in Fig. 2.

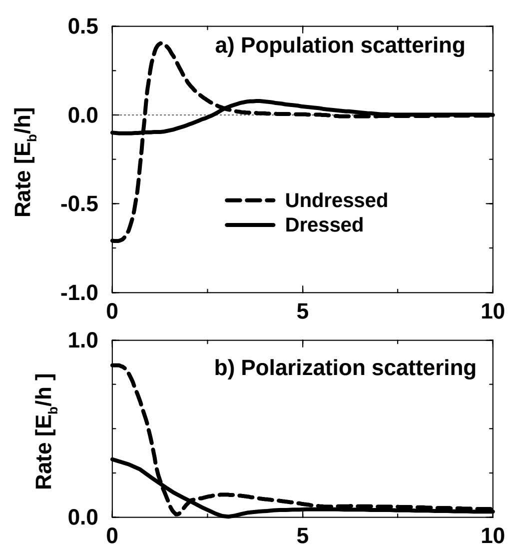

FIG. 2. Collision rates at the time indicated by the vertical arrow in Fig. 1. (a) Population scattering rate $\partial n_{1,k}/\partial t|_{\mathrm{scatt}}$ in units of E_b/h as a function of the electron-hole kinetic energy $\frac{\hbar^2k^2}{2\mu}$ (E_b units). Long-dashed line: undressed result. Solid line: dressed theory. (b) Same plot for the rate $|\partial P_k/\partial t|_{\mathrm{scatt}}|$.

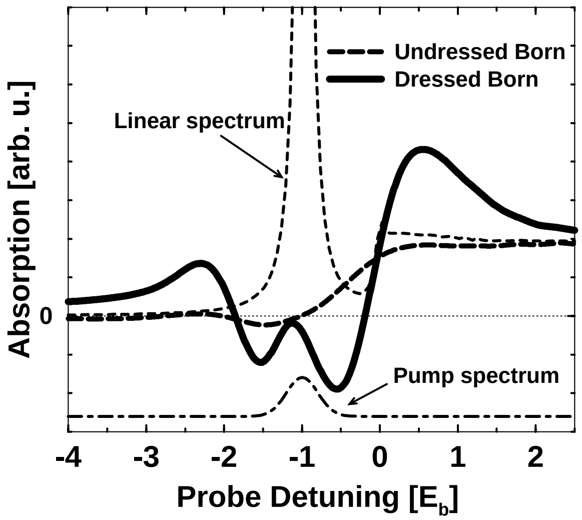

FIG. 3. Absorption coefficient (arbitrary units) versus probe detuning (E_b units). The pump-probe time delay is zero. Same parameters as in Fig. 1. Dotted-dashed line: the spectral shape of the pump beam. Short-dashed line: linear regime spectrum (no pump). Long-dashed line: undressed Born. Solid line: dressed Born.

which are in between the static and the unscreened approximation. Moreover, in a two-dimensional system, the screening is known to be less effective than in the three-dimensional case. With respect to the Markov approximation, they have found that the non-Markovian features are significant for ultrashort pump pulses. In this paper, we have considered the opposite regime of temporally large (spectrally narrow) pump beams. Therefore, deviations from the Markov approximation are expected to have a secondary role.

In conclusion, we have studied the role of Coulomb scattering in the coherent transients of strongly driven exciton resonances. Within a nonequilibrium formalism, we have derived the dressed semiconductor Bloch equations for carrier populations and interband polarization. The light-induced renormalizations are consistently included in the quasiparticle spectral functions. The light dressing is found to drastically decrease the Coulomb collision efficiency, leading to much smaller deviations from the fully coherent Hartree-Fock dynamics. Indeed, population and polarization scattering are not strong enough to prevent coherent saturation effects such as field-driven Rabi oscillations and dynamical Stark splitting.

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