Exciton Beats in GaAs Quantum Wells: Bosonic Representation and Collective Effects

J. Fernández-Rossier and C. Tejedor
Departamento de Física Teórica de la Materia Condensada,
Universidad Autónoma de Madrid, Cantoblanco, 28049 Madrid, Spain

R. Merlin
Department of Physics, The University of Michigan,
Ann Arbor, MI 48109-1120

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We discuss light-heavy hole beats observed in transient optical experiments in GaAs quantum wells in terms of a free-boson coherent state model. This approach is compared with descriptions based on few-level representations. Results lead to an interpretation of the beats as due to classical electromagnetic interference. The boson picture correctly describes photon excitation of extended states and accounts for experiments involving coherent control of the exciton density and Rayleigh scattering beating.

The optical properties of semiconductor quantum wells (QW) and, in particular, the coherent dynamics of excitons following resonant excitation with ultrafast laser pulses have attracted much attention in recent years [1–12]. It is generally accepted that there is a transfer of coherence between the optical field and the QW that disappears in a characteristic time T_2 (picoseconds for GaAs) after the laser is turned off. However, the questions as to how the coherence is actually induced and that of the nature of the coherent state of the solid are poorly understood. In this paper, we address these points by re-examining the long-standing problem of the (classical vs. quantum) nature of the ubiquitous beats associated with the light-hole (LX) and heavy-hole (HX) excitons, which are observed in transient optical experiments on QW [1–4]. To this end, we consider the coherent behavior of excitons using the simplest albeit non-trivial model where they are treated as non-interacting bosons. Hence, our work relates directly to studies for which nonlinear effects are not important [3,4,7,10–12] but it is not aimed at explaining the four-wave-mixing (FWM) experiments that dominate the field [1]. Nevertheless, since nonlinear effects are, typically, weak compared with harmonic contributions, the free-boson picture provides in all cases the correct lowest-order wavefunction of the photoexcited solid. It should be emphasized that our results apply only to excitons in *weakly-localized* states.

The structure of this paper is as follows. First, we discuss the bosonic representation of excitons in QW's, obtaining the exact collective state of a QW driven by an arbitrary laser pulse and show that its properties *vis-a-vis* coherence are identical to those of coherent optical fields [13]. From this, it follows that laser-induced coherence is a collective property of the exciton field that is not owed by individual excitons. Using the many-exciton wavefunction, we provide a quantitative description of recent experiments where two laser pulses are used to coherently control the HX density in a GaAs QW [7,10]. We also analyze the case where a single pulse excites both the LX and HX states and argue that the resulting beats [1–4,11,12] are not due to (single-exciton) quantum interference, as advocated by few-level models, but to polarization interference associated with the emission of phased arrays of classical antennas. Finally, we consider Rayleigh scattering experiments [3,4,11,12] and show that the bosonic approach accounts for the quadratic rise in the intensity at short times that is observed in the experiments [4].

At small electric fields and near band-gap excitation, the quanta of the induced polarization field, **P**, are the excitons [14]. Since their number is proportional to the illuminated volume of the sample, V, it is clear that the quantum description of an excited QW (and, as we shall see, that of the beats) becomes, in some sense, a *many-exciton* problem. Our discussion concerns itself with extended states. The atomic-like scheme [15] where excitons are represented by a collection of distinguishable few-level systems is the correct representation in the strongly localized regime as in the case of quantum dots or excitons bound to islands [16]. It should be emphasized that, in the latter picture, the optically-induced coherence relies on intra-level quantum entanglement and, therefore, that it is a *single-* or, at most, a *few-exciton* effect. For our approach to work, the areal density of photogenerated excitons, n, must be sufficiently low so that the excited state of the solid can be described as a system of non-interacting bosons [17]. Hence, our discussion applies to a GaAs QW excited with low intensity pulses using photon energies in the vicinity of the LX and HX absorption lines. Note that, under these conditions, the bosonic picture follows directly from the semiconductor Bloch equations [17] and BCS-like fermionic theories [18]. We are aware that our approach ignores all but a small fraction of the QW Hilbert space. However, the experiments considered here [3,4,7,10–12] are well described by models that rely on the same restricted basis and, thus, we conclude that the neglected sectors of the Hilbert space (*e.g.*, the electron-hole continuum) play only a secondary role in many cases of interest.

For a perfect QW (the weak-disorder limit is discussed later), the Hamiltonian describing free excitons coupled to a classical electromagnetic field is [19]:

$$\widehat{H} = \sum_{\mathbf{k},\alpha,M} \hbar\omega_{\mathbf{k},\alpha} A^{\dagger}_{\mathbf{k},\alpha,M} A_{\mathbf{k},\alpha,M} - \int \mathbf{P}\cdot\mathbf{E}(\mathbf{r},t)\, dV \quad (1)$$

where $\mathbf{E}$ is the electric field. $A^{\dagger}_{\mathbf{k},\alpha,M}$ is a *boson* operator that creates a QW exciton with in-plane momentum $\mathbf{k}$ valence band index α and pseudo angular momentum M. Relevant to our problem are the lowest-lying optically active ($M = \pm 1$) heavy- ($\alpha = H$) and light-hole ($\alpha = L$) QW states. The single-particle energy is $\hbar\omega_{\mathbf{k},\alpha} = E^g_\alpha - \epsilon_\alpha + K_E$ where $K_E = \hbar^2 k^2/2m_\alpha$ is the center-of-mass kinetic energy, m_α is the exciton mass, E^g_α is the relevant QW gap and ϵ_α is the exciton binding energy. Consider normal incidence, *i.e.*, the light couples only to the state at $\mathbf{k} = \mathbf{0}$. Using the fact that typical QW widths are considerably smaller than the light wavelength, we write the interaction term as $-V\sum_M P_M E_M$ where

$$P_M = \frac{1}{\sqrt{V}} \sum_\alpha G_{\alpha,M} \left(A^{\dagger}_{\mathbf{0},\alpha,M} + A_{\mathbf{0},\alpha,M} \right) \quad (2)$$

and $E_M(\mathbf{r}=0,t)$ are, respectively, the $M = \pm 1$ components of $\mathbf{P}$ and $\mathbf{E}$, and $G_{\alpha,M}$ are constants proportional to the dipole matrix element [17]. The Hamiltonian (1) is equivalent to that of a set of *independent* harmonic oscillators (the exciton H and L modes at $\mathbf{k} = \mathbf{0}$) driven by an external field. For arbitrary driving force and initial state, this problem can be solved exactly by applying a time-dependent Glauber transformation [13,20]. In particular, if the QW is initially in its ground state and the external field is turned on at $t = -\infty$, the exact state of the exciton field at time t is [20]

$$|\Xi\rangle = e^{-i\widehat{H}_0 t/\hbar} \prod_{\alpha,M} e^{-|K_{\alpha,M}|^2/2} e^{iK_{\alpha,M} A^{\dagger}_{\mathbf{0},\alpha,M}} |0\rangle \quad (3)$$

where $\widehat{H}_0$ is the free exciton Hamiltonian and

$$K_{\alpha,M}(t) = \frac{\sqrt{V} G_{\alpha,M}}{2\hbar} \int_{-\infty}^{t} E_M(s) e^{i\omega_{\mathbf{0},\alpha} s} ds. \quad (4)$$

The wavefunction (3) is a product of states of individual modes that is formally identical to the so-called (multimode) *coherent state* proposed by Glauber as the quantum counterpart to classical light [13]. As for the photon case, exciton coherent states are fully characterized by the complex function $K_{\alpha,M}(t)$ which defines the classical phase. Because the system is not nonlinear, the induced polarization is exactly given by

$$\langle\Xi|P_M|\Xi\rangle = \frac{2}{\sqrt{V}} \sum_\alpha G_{\alpha,M} Re\left(i e^{i\omega_{\mathbf{0},\alpha} t} K_{\alpha,M}(t) \right). \quad (5)$$

while the areal density of α-excitons with momenta $\mathbf{k}$ and M is $n_{\mathbf{k},\alpha,M} = (N_{\mathbf{0},\alpha,M} l/V)\delta_{\mathbf{k},\mathbf{0}}$where

$$N_{\mathbf{0},\alpha,M} = \langle\Xi|A^{\dagger}_{\mathbf{0},\alpha,M} A_{\mathbf{0},\alpha,M}|\Xi\rangle = |K_{\alpha,M}(t)|^2 \quad (6)$$

and l is the width of the well. Here, we note that the linear susceptibility, as easily obtained from (5), is identical to that of the analogous few-level model. These expressions apply strictly to non-interacting excitons. Coupling with the environment and interactions between excitons lead to *dephasing* in that the pure state (3) evolves into a statistical mixture of coherent states with random phases. It is beyond the scope of this work to provide a microscopic account of these interactions. Instead, we will rely on the exponential decay approximation and treat dephasing phenomenologically.

Coherent control theory.— We now analyze recent experiments where light pulses are used to control the exciton density in a GaAs-QW [7,10]. Within the bosonic description, these results constitute a striking demonstration of collective behavior. The experiments rely on two phase-locked pulses tuned close to an exciton mode of energy $\hbar\omega$ and separated by a time delay τ which serves as the control parameter for the exciton density ($n = \sum_{\mathbf{k},\alpha,M} n_{\mathbf{k},\alpha,M}$ is probed indirectly by monitoring the reflectivity of a third pulse [7] or the luminescence intensity [10]). The data can be fitted to $n = 2n_s[1 + \cos(\omega\tau)e^{-\tau/T_2}]$ where n_s represents the exciton density generated by a single pulse. Hence, small changes in the time delay ($\pi/\omega \approx 1$ fs) lead to large variations of n from zero, corresponding to destructive interference between the pulses, to nearly four times the value for one pulse [7,10]. These results can be easily explained using the expressions derived previously. To account for the double pulse, we write $E = E_1(t) + E_2(t)$ where $E_1(t) = F(t)$, $E_2(t) = F(t-\tau)$ and

$$F(t) = E_0 \sin(\Omega t) e^{-(t/T)^2}. \quad (7)$$

T and Ω are the pulse width and central frequency. From (4),(6) and (7), and assuming that the pulses couple only to a single $\mathbf{k} = \mathbf{0}$ mode of frequency ω (the extension to many modes is straightforward) we obtain the areal density

$$n \simeq |K(\infty)|^2 (l/V) = 2n_s[1 + \cos(\omega\tau)]. \quad (8)$$

$n_s = (gE_0T/\hbar)^2 \exp -[\omega^2T^2(1+r^2)/4] \sinh[r\omega^2T^2/2]$ is the average density created by one pulse, g is a constant and $r = \Omega/\omega$ measures the detuning between the laser and the exciton resonance. The result (8) contains the essential feature of coherent control, namely, the oscillatory term. Decay can be incorporated into the model by considering, e.g, a distribution of modes of different energies [21]. It should be emphasized that, in the coherent-state representation, control of the density follows from the fact that the wavefunction (3) is a linear superposition of states with different number of excitons (not surprisingly, the same result can be obtained from a classical analysis of a kicked harmonic oscillator as it is done in phonon control studies [22]). Also note that the state induced by the pair of pulses

$$|\Xi_{E_1+E_2}\rangle \sim e^{i(K_1+K_2)A^\dagger}|0\rangle \tag{9}$$

cannot be approximated by the superposition state $[1+i(K_1+K_2)A^\dagger]|0\rangle$ because $K_{1,2} \propto \sqrt{V}$. Hence, our interpretation differs significantly from that of the two-level model [7] where control is understood as a single-exciton quantum interference effect.

LX-HX beats.— We now turn to the main subject of this work, namely, the beats of frequency $\omega_L - \omega_H$ (ω_L and ω_H are, respectively, the frequencies of the LX and HX excitons) as reported in a wide variety of experiments [1–4,8], which are conventionally characterized [1,2] as a quantum interference phenomenon much like the so-called quantum beats of atomic physics [15]. Within the atomic-like interpretation, the QW is treated as a set of three-level systems whose excited states are the LX- and HX- states. The role of the optical pulses is to bring each system into a *sum* state, *i.e.*, a linear combination of LX, HX and the ground state and, thus, the beats are a consequence of intra-exciton quantum entanglement [15]. As we shall see, the actual state of the solid is very different from that of the atomic-like picture.

Consider first a perfect QW and circularly polarized pulses of bandwidth large enough so that both LX- and HX-modes of well-defined angular momentum M are resonantly excited. From (3), it directly follows that the wavefunction in this case is a *product* of LX and HX coherent states

$$|\Xi_{LH}\rangle \sim e^{-i\omega_H t N_H} e^{iK_H(t)A_H^\dagger} e^{-i\omega_L t N_L} e^{iK_L(t)A_L^\dagger}|0\rangle \tag{10}$$

where $N_\alpha \equiv A_\alpha^\dagger A_\alpha$. Using (5), it can be shown that (10) leads to LX-HX beats that reflect interference of coherent light emitted by two phased arrays of antennas. We note that the beating field, associated with $\langle\Xi_{LH}|P|\Xi_{LH}\rangle \neq 0$, is classical in nature as opposed to the field due to spontaneous emission that characterizes quantum beats [15]. Moreover, because Ξ_{LH} cannot be expressed in terms of sums of LX and HX states (since $K_{L,H} \propto \sqrt{V}$, this is not even possible as an approximation), it is apparent that quantum-superposition arguments cannot be used to describe beats associated with extended excitons.

Rayleigh scattering.— To consider the experiments on resonant secondary emission involving elastic (Rayleigh) scattering [3,4,11,12], we add to (1) the term

$$\widehat{U} = \sum_{\alpha,\mathbf{k}\neq\mathbf{0}} V_\alpha(\mathbf{k})\left(A_{\mathbf{k},\alpha}^\dagger A_{\mathbf{0},\alpha} + A_{\mathbf{k},\alpha}A_{\mathbf{0},\alpha}^\dagger\right) \tag{11}$$

describing the *elastic* scattering from the state coupled to the laser ($\mathbf{k}=\mathbf{0}$) to other states ($\mathbf{k}\neq 0$) due to interaction with defects such as impurities and interface roughness. Since the disorder described by (11) does not affect the internal degrees of freedom, our results do not apply to quantum dots [16]. $\widehat{U}$ gives rise to Rayleigh scattering, *i.e.*, emission of photons with the same energy but different in-plane momentum than the incident light [23]. Following [24], we adopt the Heisenberg picture and, according with (3), we assume that the exciton field at $t=0$ is described by $\langle A_{\mathbf{0},\alpha}\rangle = K_{\mathbf{0},\alpha}(t=0)$ and $\langle A_{\mathbf{k}\neq\mathbf{0},\alpha}\rangle = 0$ (all but the $\mathbf{k}=\mathbf{0}$ mode are empty after the pulse strikes). This approximation is valid for fast pulses and weak disorder. Since $\widehat{H}+\widehat{U}$ does not mix LX and HX, we solve for each α $(=L,H)$ the problem of a single exciton of momentum $\mathbf{k}=\mathbf{0}$ coupled to a continuum of α-excitons at $\mathbf{k}\neq\mathbf{0}$. The time evolution is given by [24]

$$\begin{aligned}\langle A_{\mathbf{k}\neq\mathbf{0},\alpha}(t)\rangle &= \frac{\Lambda_\alpha V_\alpha(\mathbf{k})}{\delta\Omega_\alpha - i\Gamma_\alpha} e^{-i\omega_\alpha t}[e^{-(i\delta\Omega_\alpha+\Gamma_\alpha)t}-1] \\ \langle A_{\mathbf{0},\alpha}(t)\rangle &= \Lambda_\alpha e^{-(i\omega_\alpha+i\delta\Omega_\alpha+\Gamma_\alpha)t}\end{aligned} \tag{12}$$

where $\Lambda_\alpha = \langle A_{\mathbf{0},\alpha}(t=0)\rangle$. $\delta\Omega_\alpha$ and Γ_α are, respectively, the small energy renormalization and the decay constant of the state at $\mathbf{k}=\mathbf{0}$ due to $\widehat{U}$. The following conclusions can be drawn from (12). First, elastic (disorder-induced) scattering leads to transfer of coherence from the mode initially excited by the laser to states with $\mathbf{k}\neq\mathbf{0}$. This accounts for the observed light emission in the non phased matched direction $\mathbf{k}\neq\mathbf{0}$. Second, within our model, the scattered field is coherent with the laser field, in agreement with recent interferometric experiments [11]. The intensity of the secondary emission is given by

$$\begin{aligned}\langle I\rangle \propto\ & \langle A_{\mathbf{k}\neq\mathbf{0},L}^\dagger\rangle\langle A_{\mathbf{k}\neq\mathbf{0},L}(t)\rangle + \langle A_{\mathbf{k}\neq\mathbf{0},H}^\dagger\rangle\langle A_{\mathbf{k}\neq\mathbf{0},H}(t)\rangle + \\ & \langle A_{\mathbf{k}\neq\mathbf{0},L}^\dagger\rangle\langle A_{\mathbf{k}\neq\mathbf{0},H}\rangle + \langle A_{\mathbf{k}\neq\mathbf{0},H}^\dagger\rangle\langle A_{\mathbf{k}\neq\mathbf{0},L}\rangle\end{aligned} \tag{13}$$

where the last two terms add up to the beating term $\cos[(\omega_L-\omega_H)t]$ observed in the experiments. It is clear that, within our model and as for the perfect QW, these Rayleigh beats are due to interference between the fields associated with the HX and LX polarizations, which behave as a distribution of *classical* antennas. Another conclusion that can be drawn from (12) is that, at short times, $\langle A_{\mathbf{k}\neq\mathbf{0},\alpha}\rangle \propto t$. This is in agreement with the *quadratic* ($\propto t^2$) rise in the Rayleigh signal observed at short times and very low exciton densities [4], which is expected when disorder is the only (or the fastest) source of scattering. The emission for $\mathbf{k}=\mathbf{0}$ decays exponentially with time constant $1/\Gamma_\alpha$ due to the coupling with the continuum of modes at $\mathbf{k}\neq\mathbf{0}$.

As discussed earlier, the perfect QW cannot be described as a collection of few-level systems because the light creates macroscopic populations of $\mathbf{k} = \mathbf{0}$ exciton modes that are uncorrelated. The situation is somehow different for $\widehat{U} \neq 0$. Here, the wavefunction can be formally obtained by applying the transformation $A_{\mathbf{k},\alpha} = \sum_{\xi} c_{\xi,\alpha}(\mathbf{k}) B_{\xi,\alpha}/\sqrt{V}$ so that the Hamiltonian $\widetilde{H}_0 = \widehat{H}_0 + \widehat{U}$ takes on the diagonal form $\widetilde{H}_0 = \sum_{\xi,\alpha} \hbar\Omega_{\xi,\alpha} B^{\dagger}_{\xi,\alpha} B_{\xi,\alpha}$ ($B_{\xi,\alpha}$ and $B^{\dagger}_{\xi,\alpha}$ are boson operators and $c_{\xi,\alpha}(\mathbf{k})$ are constants). Following the impulsive excitation, the exciton field is

$$|\widetilde{\Xi}\rangle \sim e^{-i\widetilde{H}_0 t/\hbar} \prod_{\xi,\alpha} e^{i\widetilde{K}_{\xi,\alpha}(t) B^{\dagger}_{\xi,\alpha}} |0\rangle \tag{14}$$

where $\widetilde{K}_{\xi,\alpha} = c^{*}_{\xi,\alpha}(\mathbf{0}) K_{\alpha,M}/\sqrt{V}$. As (10), $\widetilde{\Xi}$ is a product state of individual modes that carries a macroscopic *polarization* [25]. Unlike the perfect case, however, the occupation of individual modes, $\langle B^{\dagger}_{\xi,\alpha} B_{\xi,\alpha}\rangle$, is not macroscopic since $\widetilde{K}_{\xi,\alpha}$ does not depend on V; see (4). Hence, the question arises as to which extent $\widetilde{\Xi}$ can be distinguished from the sum-state

$$|\widetilde{\Psi}\rangle \sim \prod_{\xi} [1 + i\widetilde{K}_{\xi,L}(t) B^{\dagger}_{\xi,L} + i\widetilde{K}_{\xi,H}(t) B^{\dagger}_{\xi,H}] |0\rangle \tag{15}$$

that carries the same polarization and gives the same beats at $\omega_L - \omega_H$ (note that $\widetilde{\Psi}$ represents, in some sense, a collection of atomic 3-level systems with *randomly-oriented* dipole moments). The main problem with (15) is that, since the total Hamiltonian involves a sum over the light- and heavy-hole modes, the time-dependent wavefunction must be in a product form. Further, there are observable differences between states (15) and (14). The two expressions give, in general, different answers for the mode occupation which is always < 1 for $\widetilde{\Psi}$ but can have any value for $\widetilde{\Xi}$ and, moreover, the spectrum of polarization *fluctuations* $\langle P^2\rangle - \langle P\rangle^2$ evaluated with $\widetilde{\Xi}$ does not have components at $\omega_L + \omega_H$ while $\widetilde{\Psi}$ does. These differences could be used in the experiments to identify (14).

In conclusion, light-induced exciton coherence must be understood in terms of a collective description of the exciton field. This gives product as opposed to sum states. The resulting LX-HX beats are not due to quantum mechanical but to classical electromagnetic interference.

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