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Interferences and coherent control of excitons in GaAs quantum wells

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Abstract. Coherent control and light–heavy-hole beats observed in transient optical experiments in GaAs quantum wells are discussed in terms of a free-boson model. The properties of the exactly calculated wavefunction of the photoexcited system lead to a reinterpretation of the beats as due to classical electromagnetic interference, in contrast to the widely used quantum description based on few-level representations.

1. Introduction

A strong effort has been devoted recently to the coherent dynamics of quantum well (QW) excitons produced by resonant excitation with ultrafast laser pulses [1–15]. 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. The way in which the coherence is actually induced and the nature of the coherent state of the solid are poorly understood. In this paper we address the long-standing problem of the (classical versus quantum) nature of the ubiquitous beats associated with the light-hole excitons (LX) and heavy-hole excitons (HX) observed in transient optical experiments on QW [1, 3, 7, 10, 13–15]. We also address the issue of the nature of interferences which give rise to the coherent control (CC) in the two-pulse experiments [6, 12, 15]. We consider the coherent behaviour of excitons using a simple but non-trivial model where they are treated as non-interacting bosons. Therefore, our work concentrates on linear effects [6, 7, 10, 12–15] and it does not consider four-wave-mixing (FWM) experiments [1]. Nevertheless, the free-boson picture provides also for non-linear experiments the correct lowest-order description of the photoexcited QW. It must be stressed that our description applies only to excitons whose localization length is much bigger than their radius. Therefore, we assume that the disorder potential does not affect the relative-coordinate exciton wavefunction. In this case, the single-exciton few-level picture of the exciton is a phenomenological description that can lead to inconsistencies [2].

The paper is organized as follows. In section 2, we present the bosonic description of excitons in QW and compare it with the few-level picture [1, 3, 5, 6, 9]. We obtain the exact collective state of a QW driven by an arbitrary laser pulse and show that its properties are identical to those of coherent optical fields [16]. Therefore, laser-induced coherence is a collective property of the exciton field that is not owned by individual excitons. Using the many-exciton wavefunction, we derive, in section 3, various results on CC [6, 12] as well as on LX–HX beats [1, 3, 7, 10]. In particular, we provide a quantitative description of measurements where

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light is used to control the exciton density in a GaAs QW [6, 12]. In section 4, we analyse the case in which both LX and HX are excited. The resulting beats [1,3,7,10] are not due to quantum interference, as indicated by few-level models, but to classical electromagnetic interference. In section 5, we consider Rayleigh scattering and show that the bosonic approach accounts for the quadratic rise in the intensity at short times that is observed in the experiments [10].

2. Bosonic theory

Near band-gap excitation with a low electric field, the excitons are the quanta of the induced polarization field, $\boldsymbol{P}$ [17]. Their number is proportional to the illuminated volume, V, so the quantum description of an excited QW is a *many-exciton* problem. We are concerned with extended states. The atomic-like scheme [18] 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 impurities. 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-exciton* or, at most, a *few-exciton* effect. Our approximation is adequate when the areal density of photogenerated excitons, n, is sufficiently low and the excited state of the solid can be described by a set of non-interacting bosons [19]. Hence, our discussion is valid for a QW excited with low-intensity pulses using photon frequencies in the vicinity of the LX and HX absorption lines. Under these conditions, the bosonic picture follows directly from the semiconductor Bloch equations [19] and BCS-like fermionic theories [20]. Our approach ignores all but a small fraction of the QW Hilbert space. However, the experiments considered here [6, 7, 10, 12–15] are well described by models that rely on the same restricted basis and we can conclude that, in many cases of interest, the neglected sectors of the Hilbert space (e.g., the electron–hole continuum) play only a secondary role.

The coupling of free excitons to a classical electromagnetic field is described by [21]

$$\hat{H} = \sum_{k,\alpha,M} \hbar\omega_{k,\alpha} A^{\dagger}_{k,\alpha,M} A_{k,\alpha,M} - \int \boldsymbol{P}\cdot\boldsymbol{E}(\boldsymbol{r},t)\,\mathrm{d}\boldsymbol{r} \tag{1}$$

where $A^{\dagger}_{k,\alpha,M}$ is a *bosonic* operator that creates a QW exciton with in-plane momentum $\boldsymbol{k}$, valence band index α and angular momentum M. We are concerned with the lowest-lying optically active ($M = \pm 1$) heavy-hole ($\alpha = \mathrm{H}$) and light-hole ($\alpha = \mathrm{L}$) QW states. Ignoring for the moment effects due to disorder, the single-particle energy is

$$\hbar\omega_{k,\alpha} = E^{g}_{\alpha} - \epsilon_{\alpha} + \hbar^2 k^2/2m_{\alpha}$$

where m_{α} is the exciton mass, E^{g}_{α} the relevant QW gap and ϵ_{α} the exciton binding energy. $\boldsymbol{P}$ is the polarization and $\boldsymbol{E}$ is the electric field. For normal incidence the light couples only to states at $\boldsymbol{k} = 0$. Since typical QW widths are considerably smaller than the light wavelength, the interaction term is $-V\sum_M P_M E_M$ where

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

and the $E_M(\boldsymbol{r} = 0, t)$ are, respectively, the $M = \pm 1$ components of $\boldsymbol{P}$ and $\boldsymbol{E}$ and the $G_{\alpha,M}$ are constants proportional to the dipole matrix element [19]. The Hamiltonian (1) is equivalent to that of a set of *independent* harmonic oscillators (the exciton H and L modes at $\boldsymbol{k} = 0$) driven by an external field. This problem can be solved exactly for arbitrary driving force and initial state, by applying a time-dependent Glauber transformation [16, 22, 23]. 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 [23]

$$|\Xi\rangle = C\mathrm{e}^{-\mathrm{i}\hat{H}_0 t/\hbar}\prod_{\alpha,M}\mathrm{e}^{\mathrm{i}K_{\alpha,M}(t)A^{\dagger}_{\mathbf{0},\alpha,M}}|0\rangle \tag{3}$$

where C is a normalization constant [23], $\hat{H}_0$ is the free-exciton Hamiltonian and

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

The wavefunction (3) is formally identical to the (multimode) *coherent state* proposed by Glauber as the quantum counterpart to classical light [16]. The wavefunction (3) differs in an essential way from the many-exciton Fock-like wavefunction where each exciton is a linear superposition of different modes with $\boldsymbol{k} = 0$. As for the photon case, exciton coherent states are fully characterized by the complex function $K_{\alpha,M}(t)$ which defines the classical phase. Since the system is linear, the induced polarization is exactly given by

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

The density of α-excitons with momenta $\boldsymbol{k}$ and M is

$$n_{\alpha,M} = \langle\Xi(t)|A^{\dagger}_{\mathbf{0},\alpha,M}A_{\mathbf{0},\alpha,M}|\Xi(t)\rangle\frac{l}{V} = |K_{\alpha,M}(t)|^2\frac{l}{V} \tag{6}$$

where l is the width of the well. The linear susceptibility obtained from (5) is identical to that of the few-level model in the linear response approximation (always for non-interacting excitons). Exciton interaction produces *dephasing* of the pure state (3) which evolves into a statistical mixture of coherent states with random phases. To describe these interactions microscopically is beyond the scope of this work. Instead, we treat dephasing phenomenologically using an exponential decay.

3. Coherent control of the exciton density

We now analyse recent experiments where light pulses are used to coherently control the exciton density in a GaAs QW [6, 12, 24]. Within the bosonic description, these results constitute a striking demonstration of collective behaviour. 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 ($n = \sum_{\boldsymbol{k},\alpha,M} n_{\boldsymbol{k},\alpha,M}$ is probed indirectly by monitoring the reflectivity of a third pulse [6] or the luminescence intensity [12]). The data can be fitted to

$$n = 2n_s[1 + \cos(\omega\tau)\mathrm{e}^{-\tau/T_2}]$$

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 [6, 12]. These results can be easily explained using the expressions derived previously. To account for the double pulse, we write

$$E = E_0[F(t) + F(t-\tau)] = E_0[\sin(\Omega t)\mathrm{e}^{-(t/T)^2} + \sin(\Omega(t-\tau))\mathrm{e}^{-((t-\tau)/T)^2}]. \tag{7}$$

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

$$\langle n(t)\rangle = \frac{l}{V}\left|\frac{G}{2\hbar}\int_{-\infty}^{t} E_0(F(s) + F(s-\tau))\mathrm{e}^{\mathrm{i}\omega s}\,\mathrm{d}s\right|^2. \tag{8}$$

In figure 1 we plot the density of excitons as a function of time, equation (8), for four slightly different values of the delay, τ, all lying in an interval of two fs. In the figure we can see that when the second pulse arrives, the density of excitons can either go to zero (destructive interference), remain the same, increase by a factor of two or increase by a factor of 4—or display any of the possibilities in between (not displayed in the figure). The density of excitons for $t \gg \tau + T$, when the two exciting pulses are finished, dramatically depends on the value of τ:

$$n \simeq |K(\infty)|^2 (l/V) = 2n_s[1 + \cos(\omega\tau)] \tag{9}$$

where

$$n_s(\omega, \Omega, T) = (GE_0T/\hbar)^2 \exp -[\omega^2T^2(1 + r^2)/4] \sinh[r\omega^2T^2/2]$$

is the average density created by one pulse, and $r = \Omega/\omega$ measures the detuning between the laser and the exciton resonance. The result (9) contains the essential feature of CC, namely, the oscillatory term. The observed decay of the amplitude of the oscillations [6], which is not reproduced by (9), can be obtained if we include the inhomogeneous broadening in a somewhat phenomenological way by considering the following Hamiltonian:

$$\hat{H}_{IB} = \int_{-\infty}^{\infty} (D(\omega')\hbar\omega' a^{\dagger}_{\omega'} a_{\omega'} + GE(t)(a^{\dagger}_{\omega'} + a_{\omega'}))\, \mathrm{d}\omega' \tag{10}$$

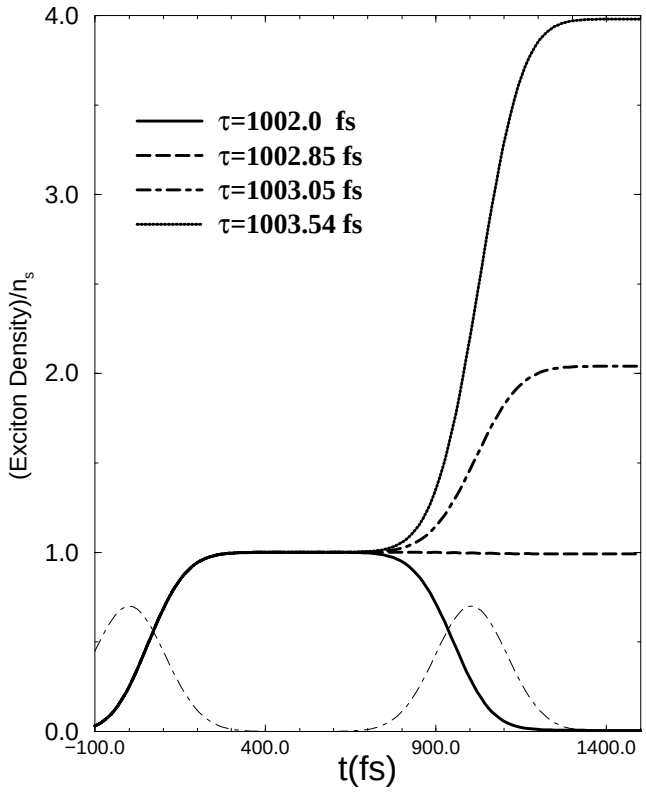

Figure 1. Exciton density, as a function of time, for four different delays τ. The laser pulse amplitudes are also plotted for reference, although their absolute value is meaningless in this figure. The position variations of the second laser pulse cannot be appreciated on the timescale of the figure.

where

$$D(\omega') \equiv (1/\Gamma)\mathrm{e}^{-((\omega'-\omega)/\Gamma)^2}$$

is the inhomogeneous distribution of excitonic energies. The analogue of equation (9) for the inhomogeneously broadened excitons is given by

$$n_{IB}(\tau, \Gamma, T) = \int_{-\infty}^{\infty} (D(\omega')2n_s(\omega', \Omega, T)(1 + \cos(\omega'\tau))) \, \mathrm{d}\omega'. \tag{11}$$

The behaviour of $n_{IB}(\tau, \Gamma, T)$ on the fs scale is identical to that of equation (9). The difference between (9) and (11) lies in the amplitude of the oscillations, which is constant in the case of (9) while it decays in the case of (11), as can be seen in figure 2. The predicted decay rate is an increasing function of the broadening. Although in this phenomenological model each excitonic mode remains fully coherent, the superposition of different energies produces the decay of the total polarization induced by the first laser pulse which in turn produces the decay of the CC. In the experiments, the decay of the polarization is also produced by any inelastic scattering undergone by the coherent excitons. This kind of decay is known as intrinsic dephasing and is a much more difficult theoretical problem. From the experimental point of view, it is not trivial to ascertain how much of the decay of the induced polarization is due to inhomogeneous broadening and how much to intrinsic dephasing.

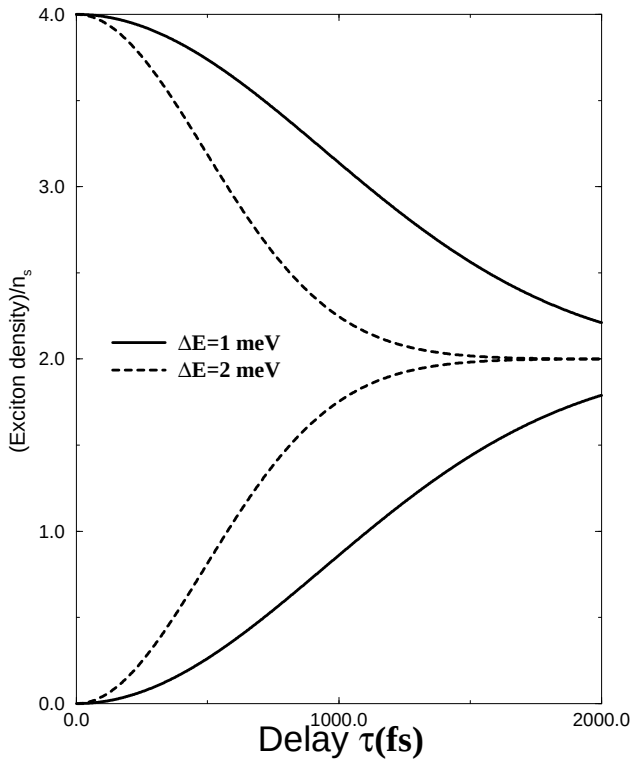

Figure 2. The maximum and the minimum exciton density for two values of $\Delta E \equiv \hbar\Gamma$ in units of the density created by a single pulse. As the delay is bigger than $1/\Gamma$, the density created by the two pulses tends to be twice the density created by a single pulse and the CC oscillations disappear.

It should be emphasized that control of the density of excitons follows from the fact that the coherent response of the system is characterized by the complex number (4) which is a

linear function of the external field. If we label the pulses $E_1(t)$ and $E_2(t)$, the complex number characterizing each excitonic mode after the interaction with the two pulses is

$$K_{tot} = |K_1|e^{i\phi_1} + |K_2|e^{i\phi_2}.$$

The density of excitons created by a single pulse, $E_{i=1,2}(t)$, is $|K_{i=1,2}|^2 \equiv |K|^2$ and cannot depend on τ. Therefore, for two identical pulses,

$$|K_{tot}|^2 = |K|^2(e^{i\phi_1} + e^{i\phi_2})^2 = 2|K|^2[1 + \cos(\phi_2 - \phi_1)].$$

In the case of a single excitonic mode, $(\phi_2 - \phi_1) = \omega\tau$. It is very important to note that the exciton wavefunction after the two pulses is *not* the sum of the wavefunctions of the system excited by a single pulse. Instead, equation (3) becomes, in the case of a single excitonic mode excited by two pulses,

$$|\Xi_{CC}\rangle \propto e^{i(K_1+K_2)A^\dagger} \propto e^{iK_1A^\dagger}e^{iK_2A^\dagger}|0\rangle \neq |\psi(E_1(t))\rangle + |\psi(E_2(t)\rangle. \tag{12}$$

This is in contrast with the microscopic CC achieved in atoms and in quantum dots [25] in which the wavefunction of the system is a *sum* of the state of the system excited only by the first pulse plus the state of the system excited only by the second pulse. In both the bosonic and the microscopic case the polarization of the system is a sum of the polarizations induced by the pulses (in the linear regime). In the microscopic case the interference can be understood as a quantum interference or a polarization interference. In the case of bosonic excitons, this is no longer true because the wavefunction is not a sum. Therefore, CC of bosonic excitons is due to the interference between the polarization induced by the first pulse and that induced by the second pulse [26]. Note that microscopic CC is essentially a single-particle effect, in contrast to bosonic CC which comes from the coherence of an assembly of excitons. In spite of these big differences, the dependences of the energy absorbed by the system, in the linear regime, as functions of the delay τ, are the same for the two cases. As the Hamiltonian (1) used in this paper is closer to the microscopic Hamiltonian than the few-level models, we think that in the case of quantum well excitons [6, 12] CC must be understood as a bosonic collective effect.

4. LX–HX beats: quantum or classical?

We now turn to analysing the beats of frequency $\omega_L - \omega_H$ with ω_L and ω_H being, respectively, the frequencies of the LX and HX excitons. These beats are reported in a wide variety of experiments [1, 3, 7, 9, 10, 13, 15], usually being characterized [1, 3] as a quantum interference phenomenon much like the so-called quantum beats of atomic physics [18]. Within the atomic-like framework, the QW is treated as a set of three-level systems whose excited states are the LX and HX states. The optical pulses bring each system into a *sum* state, i.e., a linear combination of the LX, HX and the ground state. Thus, the beats are a consequence of intra-exciton quantum entanglement [18]. However, the actual state of the solid is very different from that of the atomic-like picture and, moreover, the latter is inconsistent with measurements of secondary emission in directions other than that of the normal [7, 10, 13, 15].

A QW is photoexcited by circularly polarized pulses of bandwidth large enough that both LX and HX modes are resonantly excited. Using (3), the wavefunction is a *product* of LX and HX coherent states:

$$|\Xi_{LH}\rangle \propto e^{-i\omega_H t N_H}e^{iK_H(t)A_H^\dagger}e^{-i\omega_L t N_L}e^{iK_L(t)A_L^\dagger}|0\rangle \tag{13}$$

where $N_\alpha \equiv \langle A_\alpha^\dagger A_\alpha\rangle$. The wavefunction (13) leads to LX–HX beating as can be shown using (5). Quantum superposition arguments cannot be used to describe beats associated with extended excitons because $|\Xi_{LH}\rangle$ cannot be expressed in terms of a sum of LX and HX states. This refers to measurements involving the linear term in the expansion of $\boldsymbol{P}$ in

powers of $\boldsymbol{E}$. To understand beating in FWM experiments [1, 3], we must face the fact that optical non-linearities originate from coupling among excitons (and with other sectors of the Hilbert space). Given that interactions necessarily lead to LX–HX mixing, i.e., entanglement of fields, the question may arise as to the possibility that FWM beats (although not linear ones) could be due to atomic-like quantum entanglement, as proposed in [1, 3]. Such a scenario is largely unphysical, because $|\Xi_{\mathrm{LH}}\rangle$ is the correct lowest-order wavefunction with or without interactions. The beats observed in both linear and non-linear experiments reflect primarily coherent properties of (13). We claim that LX–HX mixing (as well as mixing with other states), while central to the question of the origin of the non-linearity, represents a small correction to $|\Xi_{\mathrm{LH}}\rangle$ and, moreover, that entanglement is only relevant to the beating phenomenon in that it allows the LX–HX correlations of (13) to manifest themselves in non-linear experiments.

5. LX–HX beats in the secondary emission

Let us consider the experiments of [7, 10], involving momentum scattering. We are going to show that the LX–HX beats reported in those experiments are due to *classical interference* of radiation emitted by the coherent exciton fields. This is supported by the fact that both the beats and $\boldsymbol{P}$ decay with the same time constant, T_2. To account for the scattering, we consider the *elastic* interaction of excitons with defects such as impurities and interface roughness. For that purpose, we must add to (1) the term

$$\hat{U} = \sum_{\alpha,\boldsymbol{q},\boldsymbol{p}} V_\alpha(\boldsymbol{q}-\boldsymbol{p})(A^\dagger_{\boldsymbol{q},\alpha}A_{\boldsymbol{p},\alpha} + A_{\boldsymbol{q},\alpha}A^\dagger_{\boldsymbol{p},\alpha}). \tag{14}$$

$\hat{U}$ gives rise to Rayleigh scattering, i.e., emission of photons with the same energy as but different in-plane momentum to the incident light [27]. Following [28], we adopt the Heisenberg picture and, in accordance with (3), we assume that the exciton field at $t = 0$ is described by $A_{\boldsymbol{0},\alpha} = K_{\boldsymbol{0},\alpha}(t = 0)$ and $A_{\boldsymbol{k}\neq 0,\alpha} = 0$ (all but the $\boldsymbol{k} = 0$ mode are empty after the pulse strikes). This approximation is valid for short pulses. Since $\hat{H} + \hat{U}$ does not couple LX and HX, we solve for each α (=L, H) the problem of a single exciton of momentum $\boldsymbol{k} = 0$ coupled to a continuum of α-excitons at $\boldsymbol{k} \neq 0$. The time evolution is exactly given by [28]

$$\begin{aligned}
\langle A_{\boldsymbol{k}\neq 0,\alpha}(t)\rangle &= \frac{\langle A_{\boldsymbol{0},\alpha}(t=0)\rangle V_\alpha(\boldsymbol{k})}{\delta\Omega_\alpha - \mathrm{i}\Gamma_\alpha}\mathrm{e}^{-\mathrm{i}\omega_\alpha t}[\mathrm{e}^{-(\mathrm{i}\,\delta\Omega_\alpha+\Gamma_\alpha)t} - 1] \\
\langle A_{\boldsymbol{0},\alpha}(t)\rangle &= \langle A_{\boldsymbol{0},\alpha}(t=0)\rangle \mathrm{e}^{-(\mathrm{i}\omega_\alpha+\mathrm{i}\,\delta\Omega_\alpha+\Gamma_\alpha)t}
\end{aligned} \tag{15}$$

where $\delta\Omega_\alpha$ and Γ_α are, respectively, the small-energy renormalization and the decay constant of the state at $\boldsymbol{k} = 0$ due to $\hat{U}$. The following conclusions can be drawn from (15).

(a) Elastic (disorder-induced) scattering leads to transfer of coherence from the mode initially excited by the laser to states with $\boldsymbol{k} \neq 0$ and to a non-zero value of $\langle\sum_\alpha P_{M,\alpha,\boldsymbol{k}}\rangle$. This accounts for emission of light in directions $\boldsymbol{k} \neq 0$ other than that of the laser pulse ($\boldsymbol{k} = 0$).

(b) The intensity of the light emitted by the excitons in directions different to that of the laser pulses (RSE) displays beats of frequency $\omega_{\mathrm{L}} - \omega_{\mathrm{H}}$, as observed experimentally [7, 10, 13–15]:

$$I_{\boldsymbol{k}} = I_{\mathrm{L},\boldsymbol{k}} + I_{\mathrm{H},\boldsymbol{k}} + 2\sqrt{I_{\mathrm{L},\boldsymbol{k}}I_{\mathrm{H},\boldsymbol{k}}}\cos[(\omega_{\mathrm{L}} - \omega_{\mathrm{H}})t] \tag{16}$$

where $I_{\alpha,\boldsymbol{k}} \propto |P_{M,\alpha,\boldsymbol{k}}|^2 \propto |A_{\boldsymbol{k},\alpha}(t)|^2$ can be obtained from (5) and (15). It is clear that, within our model, these Rayleigh beats originate from classical interference between the fields associated with the HX and LX polarizations.

(c) At short times, $A_{\alpha,k\neq 0} \propto t$. This result is in agreement with the *quadratic* ($\propto t^2$) rise in the Rayleigh scattering signal observed for short times and very low exciton densities [10]. We notice that such a behaviour is expected when disorder is the only (or the fastest) source of scattering.

(d) The RSE field is emitted by a classical source, $P_{M,\alpha,k}$, and therefore is a coherent state with a well defined relative phase with respect the exciting laser pulse [16, 29]. This is in agreement with several interferometric measurements [11, 14].

We see that the exact solution of the bosonic theory, including disorder, explains the main experimental features of the transient RSE experiments.

6. Conclusions

Our main conclusion is that the bosonic collective description of the exciton field is in agreement with several transient linear optics experiments on GaAs quantum wells. In this picture the LX–HX beats are due to classical electromagnetic as opposed to quantum mechanical LX–HX interference.

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