Restrictions on the Coherence of the Ultrafast Optical Emission from an Electron-Hole-Pair Condensate

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We report on the transfer of coherence from a quantum-well electron-hole condensate to the light it emits. As a function of density, the coherence of the electron-hole-pair system evolves from being full for the low-density Bose-Einstein condensate to a chaotic behavior for a high-density BCS-like state. This degree of coherence is transferred to the light emitted in a damped oscillatory way in the ultrafast regime. Additionally, the photon field exhibits squeezing properties during the transfer time. Our results suggest a new type of ultrafast experiments for detecting electron-hole-pair condensation.

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The generation of quantum coherence and entanglement is of much interest for testing certain aspects of the non-local predictions of quantum mechanics, as well as for applications in the emerging field of quantum information processing. The recent achievement of Bose-Einstein condensation (BEC) in dilute atomic systems [1] has triggered a great interest in looking for such quantum correlations with massive particles [2]. Furthermore, the manipulation of the interaction of light with massive particles is crucial for controlling processes which transfer coherence and/or entanglement between radiation and matter. A condensed matter system being a candidate for this goal is an electron-hole-pair condensate [3,4]. Several groups have directed their experimental efforts to produce this collective state in semiconductors [5–7]. Recently, long life-time indirect excitons, in both real and **k** spaces, have been proposed as the most robust entities towards condensation [8–10]. Hence, it is natural to inquire which signatures may be expected to be transferred from the electron-hole-pair condensate to emitted photons. The transfer of coherence is an ultrafast process that can be analyzed by means of coherent control techniques as those recently applied to semiconductor nanostructures. This work is the first step in this direction.

Coherence of a quantum system is associated with the observation of interference effects which can be described by first- and higher-order correlation functions as stated by Glauber [11]. Most of the studies of the light emitted from an electron-hole condensate that have been proposed to date rely on the lowest-order fluctuations in photon counting experiments, i.e., intensity measurements instead of the correlation functions [12,13]. It has been previously shown that at low density the emitted light should be in a coherent state [14]. However, we demonstrate here that this perfect coherence transfer is possible only asymptotically. The proper way to quantify both the amount and dynamics of transferred coherence to the radiation field is by considering the correlations between photons as a function of time [15]. Recent proposals [16] may bring this kind of photon

counting experiment within reach. The main aim of this work is to explore time-dependent higher-order coherence properties of photons emitted from a collective electron-hole pair state in an ultrafast time scale. Our results show that full coherence transfer takes a time ranging from hundred of femtoseconds to a few picoseconds. During this transfer time squeezing of photons may be achieved.

Since we are interested in the quantum effects we restrict ourselves to study ground-state properties (zero temperature) and let the pair density change. We consider indirect semiconductor quantum wells where electrons and holes are spatially separated by an interlayer distance d. The confinement in the z direction is sufficiently strong so that we ignore excitations in that direction. The Hamiltonian of the system includes the kinetic energy and all the interactions among electrons and holes, the free (quantized) electromagnetic field, and the interaction between the radiation and the electron-hole system. The system's initial state is assumed to be a product of an empty radiation field state and a condensate electron-hole state, $|0_{\text{photons}}\rangle \times \prod_{\vec{k}}(u_{\vec{k}} + v_{\vec{k}} e_{\vec{k}}^{\dagger} h_{-\vec{k}}^{\dagger})|0\rangle$ with $\vec{k}$ being a two-dimensional wave number vector, $u_{\vec{k}}$ and $v_{\vec{k}}$ satisfy the normalization condition $u_{\vec{k}}^2 + v_{\vec{k}}^2 = 1$. $e_{\vec{k}}^{\dagger}$ ($h_{\vec{k}}^{\dagger}$) is the electron (hole) creation operator and $|0\rangle$ denotes the semiconductor ground state. The variational BCS-like function has enjoyed considerable success in the description of stationary properties of electron-hole systems [17]. It captures the essential electron-hole pairing correlations in the low as well as high pair density, n, limits, although it does not describe possible collective modes in the condensate. We also assume that in the ultrafast coherence transfer period just a few bunches of photons are emitted, so that n does not change significantly and thus we take it as a constant. The time-dependent optical coherence is obtained via photon operators determined by pairs evolving under the action of the electron-hole Hamiltonian in the free Bogoliubov quasiparticles approximation [18] (quantum fluctuation terms are neglected). These basic assumptions are indeed satisfied for transfer times below a few picoseconds

and more interestingly they also yield to the correct stationary limit. Coefficients $u_{\vec{k}}$ and $v_{\vec{k}}$ are found from the self-consistent solution of the BCS gap equation for given chemical potential μ and n. In what follows, energy is measured in 2D Rydbergs $\mathcal{R}_0$, length in 2D Bohr radius a_0, and time in units of $\tau_0 = \mathcal{R}_0^{-1}$.

Since an electron-hole condensate corresponds to a coherent macroscopic polarization of the electronic states, we start by considering the polarization correlation functions (PCFs). The first- and second-order PCFs are defined as $G_P^{(1)}(t_1; t_2) = \langle P_0^{\dagger}(t_1) P_0^{\dagger}(t_2) \rangle$ and $G_P^{(2)}(t_1; t_2) = \langle P_0^{\dagger}(t_1) P_0^{\dagger}(t_2) P_0^{\dagger}(t_2) P_0^{\dagger}(t_1) \rangle$, respectively, where $P_{\vec{q}} = \sum_{\vec{k}} h_{\vec{q}-\vec{k}} e_{\vec{k}}$ is the polarization field. Because of the long wavelength of emitted photons, only $\vec{q} = \vec{0}$ modes are important. PCFs in the BCS state are better expressed in terms of the so-called normal and anomalous (or pair) Green's functions [15], defined by $\mathcal{G}(t_1 - t_2) = \sum_{\vec{k}} \langle e_{\vec{k}}^{\dagger}(t_1) e_{\vec{k}}(t_2) \rangle \langle h_{-\vec{k}}^{\dagger}(t_1) h_{-\vec{k}}(t_2) \rangle$ and $\mathcal{F}(t_1 - t_2) = \sum_{\vec{k}} \langle h_{-\vec{k}}(t_1) e_{\vec{k}}(t_2) \rangle$, respectively. The first-order PCF can be written as $G_P^{(1)}(t_1 - t_2) = \mathcal{G}(t_1 - t_2) + |\mathcal{F}(0)|^2$. It should be noted that the polarization field remains always in a stationary state in such a way that its correlation and coherence functions are dependent only on $\tau = t_2 - t_1$ but not on $T = (t_1 + t_2)/2$. The first-order polarization coherence function is expressed as

$$g_P^{(1)}(\tau) = \frac{G_P^{(1)}(\tau)}{G_P^{(1)}(0)} = \frac{\sum_{\vec{k}} |v_{\vec{k}}|^4 e^{iE_{\vec{k}}\tau} + [\sum_{\vec{k}} u_{\vec{k}} v_{\vec{k}}]^2}{\sum_{\vec{k}} |v_{\vec{k}}|^4 + [\sum_{\vec{k}} u_{\vec{k}} v_{\vec{k}}]^2} , \quad (1)$$

where $E_{\vec{k}}$ is the excitation energy of Bogoliubov quasiparticles. Similarly, the second-order polarization coherence function is

$$g_P^{(2)}(\tau) = 1 + |g_P^{(1)}(\tau)|^2 - \frac{|\mathcal{F}(0)|^4}{|G_P^{(1)}(0)|^2} , \quad (2)$$

where $|\mathcal{F}(0)|^2 = [\sum_{\vec{k}} u_{\vec{k}} v_{\vec{k}}]^2$. It must be stressed that the second-order coherence function for the polarization, $g_P^{(2)}(0)$, equals 1 for a coherent state, whereas it is 2 for a chaotic field [19].

Our results for the second-order polarization field coherence, for $d = 0$ and different densities, are shown in Fig. 1. Clearly, a full coherent behavior, in the Glauber sense, appears in the low-density limit. In this case $v_{\vec{k}}^2 \ll 1$, yielding to $|g_P^{(1)}(\tau)| = |g_P^{(2)}(\tau)| \simeq 1$, as expected for an ideal electron-hole BEC. By contrast, as the density increases $g_P^{(2)}(0)$ becomes greater than 1, going up to a maximum value of 2. As a function of τ, $g_P^{(2)}$ decreases from its initial value in an oscillatory way, with a period determined by $E_{\vec{k}=0}$. At high densities $g_P^{(1)}(\tau)$ decays to zero and $g_P^{(2)}(\tau)$ decays to one in a faster oscillating way with a frequency approaching that of the system's Fermi energy. In the very-high-density limit, where $v_{\vec{k}} = 1$ for k smaller than the Fermi wave vector $\vec{k}_F$ and 0 otherwise, the anomalous term vanishes yielding to $g_P^{(1)}(\tau) = $
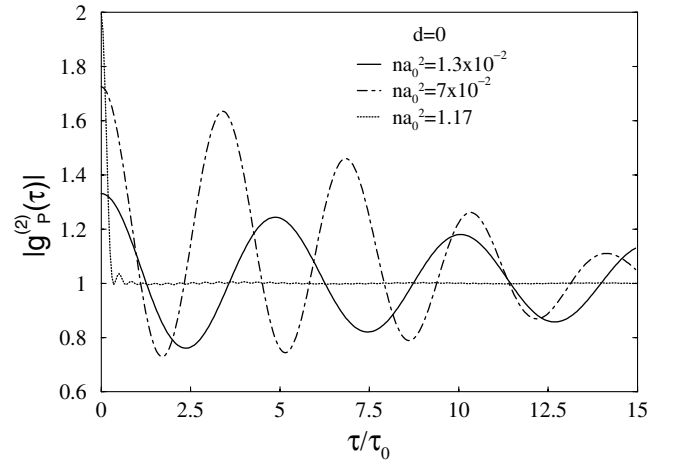

FIG. 1. Second-order polarization field coherence as a function of τ for $d = 0$ and different densities.

$[e^{-i\epsilon_F \tau} \sin(\epsilon_F \tau)]/\epsilon_F \tau$ and $g_P^{(2)}(\tau) = 1 + |g_P^{(1)}(\tau)|^2$ with $\epsilon_F = k_F^2/2$. Clearly, $g_P^{(2)}(0) = 2$, corresponding to a noninteracting fermion field, and it decays, as a function of τ, in an overdamped way.

Next we discuss the coherence properties of the emitted light in order to study the transfer of coherence from the condensate. First- and second-order correlation functions are $G_{\vec{q}}^{(1)}(t_1; t_2) = \langle C_{\vec{q}}^{\dagger}(t_1) C_{\vec{q}}(t_2) \rangle$ and $G_{\vec{q}}^{(2)}(t_1; t_2) = \langle C_{\vec{q}}^{\dagger}(t_1) C_{\vec{q}}^{\dagger}(t_2) C_{\vec{q}}(t_2) C_{\vec{q}}(t_1) \rangle$, where $C_{\vec{q}}^{\dagger}$ ($C_{\vec{q}}$) is the photon creation (annihilation) operator. For $\vec{q} = 0$ photons, $G^{(1)}(t_1; t_2) = |M_o|^2 e^{-i\omega_0(t_2-t_1)} \int_0^{t_1} dt_a \int_0^{t_2} dt_b \, f(t_b - t_a)$ with $f(t_b - t_a) = e^{i(\omega_0 - E_g - \mu)(t_b - t_a)} G_P^{(1)}(t_b - t_a)$, where ω_0 is the frequency at which the emitted light is filtered, E_g is the energy gap, and M_0 is the coupling between the photons and the electron-hole pairs. Similarly, $G_{\vec{q}}^{(2)}(t_1; t_2)$ is closely related to $G_P^{(2)}(t_1; t_2)$. From these relations between polarization and radiation correlation functions the coherence transfer from the former to the latter can be obtained. The first-order correlation function can be written as $G^{(1)}(T, \tau) = N(T, \tau) + A(T, \tau)$, where the normal and the anomalous dimensionless contributions are given by

$$N(T, \tau) = 2M_0^2 e^{-i\omega_0 \tau} \sum_{\vec{k}} |v_{\vec{k}}|^4 e^{-i(\omega + E_k)\tau}$$
$$\times \left[\frac{\cos[(\omega + E_k)\tau/2] - \cos[(\omega + E_k)T]}{(\omega + E_k)^2} \right] \quad (3)$$

and

$$A(T, \tau) = 2M_0^2 |\mathcal{F}(0)|^2 e^{-i(\omega + 2\omega_o)\tau/2}$$
$$\times \left[\frac{\cos(\omega \tau/2) - \cos(\omega T)}{\omega^2} \right], \quad (4)$$

respectively, with $\omega = \omega_0 - E_g - \mu$. The first- and second-order coherence functions are then

$$g^{(1)}(T,\tau) = \frac{G^{(1)}(T,\tau)}{\sqrt{G^{(1)}(t_1;t_1)G^{(1)}(t_2;t_2)}} \qquad (5)$$

and

$$g^{(2)}(T,\tau) = 1 + |g^{(1)}(T,\tau)|^2 - \frac{|A(T,\tau)|^2}{G^{(1)}(t_1;t_1)G^{(1)}(t_2;t_2)}, \qquad (6)$$

respectively. As for the polarization, the anomalous contribution determines whether or not the light is second-order coherent. The steady-state expressions can be quickly obtained by letting $T \gg \tau$ which leads to $G^{(1)}(t_1;t_1) \simeq G^{(1)}(t_2;t_2) \simeq G^{(1)}(T,\tau=0)$.

First- and second-order coherence do not depend on the efficiency of the detector used in photon counting experiments nor on the coupling M_0. Moreover, the amount of coherence depends only on the system's density. Turning this around, we can use the coherence information to determine the electron-hole density.

One expects the emitted light to be fully coherent in the low-density limit whereas it should be chaotic in the high-density limit as emitted by an uncorrelated source. We indeed show that in both cases the asymptotic exact solutions are recovered. Obviously, from Eq. (5) it can be drawn that $g^{(1)}(T,0) = 1$ for any frequency and/or electron-hole pair density, in agreement with the standard result according to which any single mode radiation field is first-order coherent [19]. Therefore, we restrict ourselves to second-order coherence properties. In the low-density limit, v_k is essentially the hydrogenic ground state wave function, $v_k \simeq \sqrt{n}\,\phi_{1s}(k) = [2\sqrt{2\pi n}\,a_0]/[1 + k^2 a_0^2]^{3/2}$, $\mu \simeq -\mathcal{R}_0$, and the normal contribution vanishes. The second-order coherence function becomes $|g^{(2)}(T,\tau)| \simeq 1$, indicating the fact that in a steady-state situation the light emitted from an electron-hole pair condensate (BEC state) is second-order coherent, in full agreement with previous results for excitons [14]. In the very-high-density limit, where $\mu \simeq k_F^2$, the anomalous part goes to zero. The second-order coherence is now given by $g^{(2)}(T,\tau) = 1 + |g^{(1)}(T,\tau)|^2$, so that $g^{(2)}(T,0) \simeq 2$ as it corresponds to chaotic radiation [20].

Now we turn to the more general case of arbitrary densities. Figure 2 depicts $g^{(2)}(T,0)$ at $\omega_0 = E_g + \mu$, for different densities. Clearly, two very different behaviors are observed depending on the time scale considered. In the short time regime, the emitted radiation is *partially coherent* since $g^{(2)} > 1$; this behavior is reinforced as the density increases. For $T \to 0$, $g^{(2)}(T,0)$ approaches $g_P^{(2)}(0)$, which depends only on the system's density and characterizes the fluctuations of the macroscopic collective polarization state. In this way emitted photons could bring well differentiated information on the system's ground state.

Second-order coherence describes also the tendency of photons to arrive in pairs ($g^{(2)}(T,0) > 1$) or rather to be spaced out in time ($g^{(2)}(T,0) < 1$) [19]. Our results show that at a short time scale the first condition is satisfied,
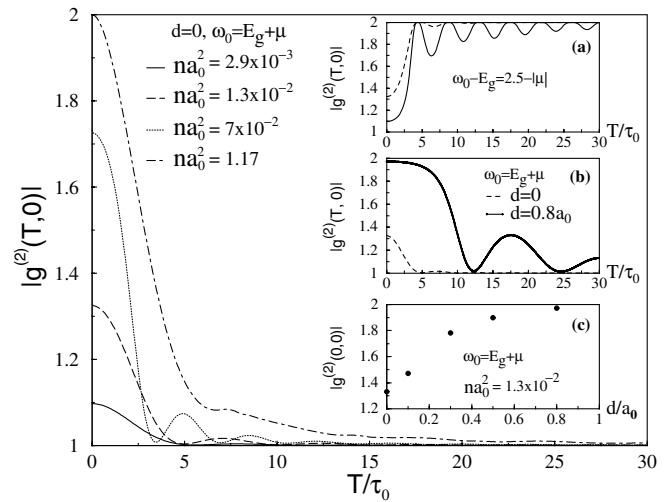

FIG. 2. Second-order coherence as a function of T for light emitted at $\omega_0 - E_g = \mu$ and $d = 0$ for different densities. Insets: (a) $g^{(2)}(T,0)$ at $\omega_0 - E_g = -2.5\mu$; (b) $g^{(2)}(T,0)$ for two different d values, and (c) $g^{(2)}(0,0)$ for $na_0^2 = 1.3 \times 10^{-2}$ as a function of the electron-hole layer separation distance.

producing photon bunching. In a long time scale, the photon bunching effect disappears and the radiation field becomes *asymptotically coherent,* i.e., $g^{(2)}(T,0) \to 1$, for any finite density. This is due to the fact that the normal contribution, $N(T,0)$, saturates to a constant value while the anomalous contribution, $A(T,0)$, grows as T^2 [20]. It must be stressed that the light emitted is coherent even though the polarization field is incoherent in the Glauber sense. The time to reach the steady-state value is longer as the density increases. For systems of interest, (i) GaAs, $\mathcal{R}_0 \simeq 16$ meV, $a_0 \simeq 62.5$ Å, and for a density 3×10^{10} cm^{-2}, the stationary regime is reached roughly after 1 ps; (ii) CdS, $\mathcal{R}_0 \simeq 120$ meV, $a_0 \simeq 12.75$ Å, and for a density 7×10^{11} cm^{-2} a steady-state situation is reached for a time on the order of 100 fs. In both cases, $na_0^2 = 1.3 \times 10^{-2}$.

These results show how the coherence of the photon field at $\omega_0 - E_g = \mu$ evolves from a partially coherent behavior, dominated by the fermionic character of the system, towards a full coherent behavior, reflecting the system's macroscopic quantum properties. By contrast, light observed at frequencies such that $\omega_0 - E_g < \mu$ evolves as a function of time from a partially coherent character towards a full chaotic behavior, i.e., $g^{(2)} \to 2$ (Fig. 2 inset (a); note that for plotted densities $\mu < 0$). Clearly, a successful coherence transfer is possible only for the former case but not for the latter one.

Interlayer separation effects are shown in insets (b) and (c) of Fig. 2. For increasing d the incoherence of light in the ultrafast regime becomes more evident and the evolution of $g^{(2)}(T,0)$ towards its coherent value is slower. For a fixed density, $g^{(2)}(T,0)$ when $T \simeq 0$, as a function of d, saturates to a final value of 2, indicating an enhancement of the chaotic behavior of light.
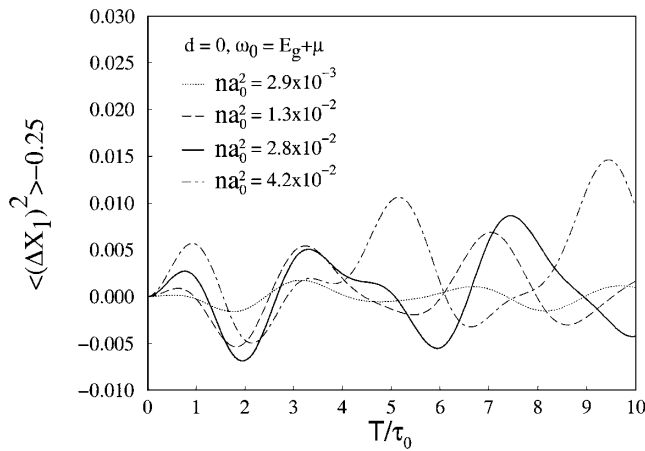
FIG. 3.   Time evolution of the photon field amplitude variance for radiation emitted at $\omega_0 = E_g + \mu$ and $M_0 = 0.1\mathcal{R}_0$.

In order to further characterize the statistical properties of the emitted radiation, we calculate the variance of the photon field amplitudes $\hat{X}_1 = \frac{1}{2}\{C(T) + C^\dagger(T)\}$ and $\hat{X}_2 = \frac{1}{2i}\{C(T) - C^\dagger(T)\}$. For radiation emitted by the condensate, these variances are $\langle(\Delta\hat{X}_{1,2})^2\rangle = \frac{1}{4} + \frac{1}{2}N(T,0) \pm \frac{1}{2}\,\mathrm{Re}[\langle\Delta C(T)^2\rangle]$ where, for $\omega_0 = E_g + \mu$, $\langle\Delta C(T)^2\rangle = -2M_o^2 e^{-i2\mu T}\sum_{\vec{k}} u_k^2 v_k^2 \frac{\sin^2(E_k T/2)}{E_k^2}$. In contrast to $g^{(1)}$ and $g^{(2)}$, these variances depend on the coupling M_0. Clearly, squeezed light is possible only at moderate low densities, where the normal contribution is negligible but $u_k v_k$ is still important. This nonlinear effect is due to interactions between electron-hole pairs, in agreement with results obtained by a simple interacting boson model [21]. Figure 3 displays the deviation of $\langle(\Delta\hat{X}_1)^2\rangle$ from $1/4$ (the coherent state value) for different densities. The amount of squeezing, as measured by the most negative value for each curve in Fig. 3, is a nonmonotonic function of the pairs density. There is a maximum squeezing in one of the quadratures every time $\mu T = \pi$.

In summary, we have shown how coherence transfer, from an electron-hole condensate to the photons it emits, proceeds as a function of time. The condensate itself presents different degrees of Glauber coherence depending on its density and the electron-hole layer separation. A full coherence transfer is restricted to light with a frequency given by $\omega_0 = E_g + \mu$ and for times greater than a few hundred femtoseconds. We also predict light squeezing from a moderate low-density electron-hole-pair system. These coherence transfer properties should help experimentalists searching for evidences of electron-hole-pair condensation in quantum wells.

[1] M. H. Anderson *et al.,* Science **269**, 198 (1995); K. B. Davis *et al.,* Phys. Rev. Lett. **75**, 3969 (1995).
[2] *Bose-Einstein Condensation,* edited by A. Griffin, D. W. Snoke, and S. Stringari (Cambridge University Press, Cambridge, 1995).
[3] L. V. Keldysh and Y. V. Kopaev, Fiz. Tverd. Tela (Leningrad) **6**, 2791 (1964) [Sov. Phys. Solid State **6**, 2219 (1965)].
[4] J. M. Blatt *et al.,* Phys. Rev. **126**, 1691 (1962).
[5] D. W. Snoke *et al.,* Phys. Rev. B **41**, 11 171 (1990).
[6] T. Fukazawa *et al.,* Phys. Rev. Lett. **64**, 3066 (1990).
[7] L. V. Butov *et al.,* Phys. Rev. Lett. **73**, 304 (1994).
[8] L. V. Butov *et al.,* Phys. Rev. B **62**, 1548 (2000).
[9] Yu. E. Lozovik and I. V. Ovchinnikov, Solid State Commun. **118**, 251 (2001).
[10] L. V. Butov *et al.,* Phys. Rev. Lett. **86**, 5608 (2001).
[11] R. J. Glauber, Phys. Rev. **131**, 2766 (1966).
[12] Th. Ostreich *et al.,* Solid State Commun. **100**, 325 (1996).
[13] T. J. Inagaki *et al.,* Solid State Commun. **115**, 645 (2000).
[14] J. Fernandez-Rossier *et al.,* Solid State Commun. **108**, 473 (1998).
[15] B. Laikhtman, Europhys. Lett. **43**, 53 (1998).
[16] P. Michler *et al.,* Nature (London) **406**, 968 (2000).
[17] C. Comte and P. Nozieres, J. Phys. (Paris) **43**, 1069 (1982).
[18] C. Comte and G. Mahler, Phys. Rev. B **34**, 7164 (1986).
[19] R. Loudon, *The Quantum Theory of Light* (Oxford University Press, New York, 2000), 3rd ed.
[20] A. Olaya-Castro, Master's thesis, Universidad de los Andes, 2001 (unpublished).
[21] Nguyen Ba An, Phys. Rev. B **48**, 11 732 (1993).