Exciton condensates in semiconductor quantum wells emit coherent light

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(February 1, 2008)

We show that a quasi-two dimensional condensate of optically active excitons emits coherent light even in the absence of population inversion. This allows an unambiguous and clear experimental detection of the condensed phase. We prove that, due to the exciton-photon coupling, quantum and thermal fluctuations do not destroy condensation at finite temperature. Suitable conditions to achieve condensation are temperatures of a few K for typical exciton densities, and the use of a pulsed, and preferably circularly polarized, laser.

PACS numbers: 71.35.+z

A system of bosons or paired fermions is said to be collectively condensed when it exhibits correlations that reflect the macroscopic occupation of a single microscopic state. Condensation can be described in terms of a non-zero order parameter corresponding to the thermal average of an operator that creates either a boson or a pair of fermions [1–3]. In the condensed phase, the system is, locally, in a coherent superposition of eigenstates with a varying number of particles. [3] Condensation has been clearly established for superconductors and superfluid ^{3}He in the case of pairs of fermions, and for superfluid ^{4}He and atomic Bose condensation in the case of bosons.

The condensation of an exciton gas in semiconductors has been extensively discussed in the literature. [1,4–18] The transition takes place when the system meets the so-called quantum-degeneracy criterion $n\lambda^D > 1$. Here, $\lambda = \sqrt{2\pi\hbar^2/Mk_BT}$ is the De Broglie thermal wavelength, n is the particle density, D is the number of spatial dimensions, and M is the particle mass [1]. Thus, the criterion is met for small M at low T and high n. Apart from $n\lambda^D$, another relevant dimensionless quantity for excitonic systems is na^D where a is the exciton radius. For $na^D << 1$ excitons behave like weakly interacting bosons, otherwise they must be treated as pairs of fermions. [4,6,7] For $n\lambda^D > 1$, condensation in the exciton gas can be expected in two flavors depending on the exciton size: a genuine Bose condensation for $na^D << 1$ and, in the oppoiste limit, a BCS-like transition as for superconductors or ^{3}He. The finite lifetime of excitons represents an obvious drawback to achieve condensation. However, we note that typical lifetimes are long enough for the excitons to reach a quasi thermodynamic equilibrium, which can be studied using time-resolved techniques. [19]

The condensation of optically active (OA) excitons has received considerable experimental [8,9] and theoretical [4–8,11,12,14–16,18] attention in the past several years. In such a system a crucial question arises as to the character of the light emitted by the condensate. Superradiance and coherent emission have been considered in the past [8,11,14,15,18] but, to the best of our knowledge, a definitive discussion of emission properties has not been given before [13].

In this letter, we consider the light emitted from a condensate of OA excitons, created using a fast laser pulse, in semiconductor Quantum Well (QW) structures. Irrespective of the process that drives the excitons to condense after the laser is turned off, we prove that: (i) the emitted light is a coherent electromagnetic field (EMF) of the Glauber type [20]; (ii) condensation of OA excitons can take place at finite temperatures in an infinite and flat two-dimensional (2D) system. This is significantly different from the case of an *isolated* 2D system for which Hohemberg theorem [21] forbids condensation at finite temperatures. The importance of (i) is twofold. First, the coherent emission could be used to probe exciton condensation . Second, the emission from the condensate is expected to occur at any density even if there is no population inversion [15]. Therefore, it constitutes a physical phenomenon different from that of lasing in high density exciton gases where coherent emission follows from population inversion. [12]

We are interested in a semiconductor QW excited by a laser pulse with a central frequency that is resonant with the lowest absorption exciton peak. The laser excitation leads to two related effects in that it creates an exciton gas in the QW, and it also induces a time dependent electric dipole moment $\mathbf{P}$. The latter results from the fact that the resonantly excited QW is in a many-exciton coherent quantum state. [22] After the laser pulse has passed, the excited QW system will require a certain time to attain equilibrium. Let us discuss first the normal case in which there is no condensation. Here, we need to consider two relaxation processes. The faster one is the decay of the laser induced

$\langle \mathbf{P} \rangle$ due to exciton thermalization: the exciton coherent state is replaced by a thermal distribution. This is strongly supported by experiments indicating that, a short time after the transit of the laser pulse, the exciton gas can be described in terms of an equilibrium distribution [7,19,22]. In this context it makes sense to characterize the exciton gas by a temperature T_X, and by a density n, that is proportional to the electric dipole fluctuation $\langle |\mathbf{P}|^2 \rangle$. The slower relaxation process is the exciton recombination producing the decay of n. This kind of relaxation is slow enough so that the excitons can be considered to be, at any time, in thermal equilibrium. Exciton recombination gives rise to incoherent light emission or luminescence, coming from thermal and quantum fluctuations of $\mathbf{P}$ around its zero mean value. A completely different scenario occurs when excitons condense because, as shown below, the electric dipole moment is non zero even in the absence of an external EMF. In this case, photon emission has a coherent component originating in the oscillating electric dipole moment. Since the emission from the condensate $\mathbf{P}$, can be mistaken for that of the external laser it is desirable to perform the condensation experiments with a pulsed source.

In the following, we show that a condensate of 2D excitons gives rise to coherent light emission. We focus on 2D systems (as a GaAs QW) in order to avoid a discussion of polariton effects, that are important in bulk semiconductors. [22] However, an oscillating electric dipole should also appear in a three dimensional condensate, resulting in coherent emission for samples of dimensions comparable to the light wavelength.

The Hamiltonian of the photons plus the QW excitons is $H = H_X + H_L + H_{XL}$ where

$$H_X = T_e + T_h + V_{ee} + V_{hh} + V_{eh} \tag{1}$$

$$H_L = \sum_{\mathbf{q},m} \hbar c q b^\dagger_{\mathbf{q},m} b_{\mathbf{q},m}, \tag{2}$$

$$H_{XL} = g D_{cv} \sum_{\mathbf{k},\mathbf{q},m} c^\dagger_{\mathbf{k},\sigma_e} d^\dagger_{\mathbf{k},\sigma_h} b_{\mathbf{q},m} + h.c. \tag{3}$$

The exciton component of the Hamiltonian, H_X, contains all the kinetic (T's) and Coulomb interaction (V's) terms corresponding to electrons and holes created by the operators $c^\dagger_{\mathbf{k},\sigma_\mathbf{e}}$ and $d^\dagger_{\mathbf{k},\sigma_\mathbf{h}}$ where $\mathbf{k}$ is the fermion 2D wavevector and the σ's are the third components of the fermion angular momenta. $b^\dagger_{\mathbf{q},\mathbf{m}}$ are the photon operators with polarization $m = \sigma_e + \sigma_h$ (OA excitons) and momentum $\mathbf{q}$ along the direction perpendicular to the QW due to conservation of in-plane momentum in Eq. (3). H_{XL} is the exciton-light dipolar coupling given in the rotating wave approximation. H_{XL} gives processes in which one exciton is created and one photon is destroyed or vice versa; the total number of excitons *plus* photons is conserved. g is the light-matter coupling constant [22] and D_{cv} is the matrix element of the dipole moment operator between the valence and the conduction band. By definition, $D_{cv} \neq 0$ in an OA semiconductor. We also impose that D_{cv} is real and we ignore its dependence on $\mathbf{k}$. [22] In GaAs QW structures, we need to consider two classes of OA excitons characterized by the third component of its total angular momentum m. Pumping by circularly polarized (left or right) light selects $m = \pm 1$. Using a mean-field approach which neglects electron-hole exchange, it has been shown [6,16] that excitons with $m = 1$ and $m = -1$ do not couple to each other. This allows us to consider two independent coexisting exciton gases each one coupled to a different light polarization state.

The state of exciton condensation is defined as the emergence, without any external EMF, of the complex order parameter: [1–4,6,7,11]:

$$\langle c^\dagger_{\mathbf{k},\sigma_\mathbf{e}} d^\dagger_{\mathbf{k},\sigma_\mathbf{h}} \rangle \equiv f_{\mathbf{k},\mathbf{m}} e^{-i\phi_m} \tag{4}$$

where $\langle\rangle$ stands for the statistical average. An essential feature of the condensation is that the phase ϕ_m of the order parameter is independent of $\mathbf{k}$, i.e., the phase is the same across the exciton gas, a signature of long range order. The magnitude of the order parameter $f_{\mathbf{k},\mathbf{m}}$ can be calculated in two different situations. In the mean field BCS approximation, valid for arbitrary densities, $f_{\mathbf{k},\mathbf{m}} = u^m_\mathbf{k} v^m_\mathbf{k}$ where $u^m_\mathbf{k}$ and $v^m_\mathbf{k}$ are the coefficients of a BCS wave function to be calculated for each particular problem. [11,16] In the limit of low densities, excitons behave like nearly free bosons and, thus, $f_{\mathbf{k},\mathbf{m}} = \sqrt{N_m}$, where N_m is the number of excitons of type m [1–3]. Our results do not depend on the approximations made to calculate $f_{\mathbf{k},\mathbf{m}}$.

Let us now discuss the behavior of the electric dipole moment in terms of its circular components $\mathbf{P}_m = \mathbf{P}_x + im\mathbf{P}_y$ with $m = \pm 1$ which couple to photons. They are given by [22]

$$P_m \equiv \sum_{\mathbf{k},\mathbf{m}} D_{cv} c^\dagger_{\mathbf{k},\sigma_\mathbf{e}} d^\dagger_{\mathbf{k},\sigma_\mathbf{h}} + h.c. \tag{5}$$

Combining Eqs. (4) and (5), we get that, for an m type exciton condensation, the mean value of P_m is non zero:

$$\langle P_m \rangle = F(N_m) cos(\phi_m) \tag{6}$$

where $F(N_m) = 2D_{cv} \sum_{\mathbf{k}} f_{\mathbf{k},\mathbf{m}}$. It follows that exciton condensation implies that $\langle P \rangle$ is non zero with no external EMF. In the presence of an external EMF, the response function of the condensate exhibits a singular behavior. [5] In the normal case, the many exciton quantum states are assumed to be eigenstates of the exciton number operator and, therefore, both the condensation order parameter and $\langle P \rangle$ are equal to zero.

We now show that the condensation-induced $\mathbf{P}$ oscillates with a frequency identical to that of the lowest exciton state This oscillating dipole emits coherent light. A full quantum mechanical calculation of the time evolution of $\mathbf{P}$ is given in reference [22]. The same result can be obtained in a simpler way from the equation of motion of the phase given by [3]

$$\frac{d}{dt}\phi_m = \hbar^{-1} \frac{\partial}{\partial N_m} E(N_m, \phi_m) \tag{7}$$

where $E(N_m, \phi_m) = \langle H_X + H_{XL} \rangle$. There are three contributions to $E(N_m, \phi_m)$. From large to small, we have the single exciton contribution $(E_g - E_b)N_m$ where E_g is the semiconductor gap and E_b is the binding energy, the exciton-exciton interaction and the exciton-light contribution $\langle H_{XL} \rangle$. In a GaAs QW, the first one is approximately 1.5 ev giving rise to an electric dipole oscillation period in the scale of femtoseconds. The many-exciton corrections simply give a renormalization $\overline{E}_b - E_b$ of the exciton binding energy of the order of 1meV. [7,11,16]

Finally, the exciton-light interaction which gives a contribution proportional to $cos(\phi_m)$; this term is negligible compared with the single exciton term because g is typically three orders of magnitude smaller than the gap. In contrast to the phase dynamics, $f(N_m)$ is a very slow quantity because the density varies in a ps scale. [19] Hence, we have a time dependent electric dipole moment

$$\langle P_m(t) \rangle = F(N_m(t)) cos\left(\frac{(E_g - \overline{E}_b)t}{\hbar} + \phi_m(0) \right) \tag{8}$$

which leads to an oscillating EMF. Our next task is to provide a quantum description of the emission process and show that the photon field emitted by the condensate is in a quantum mechanical coherent state. [20] We only consider the emission resulting from the condensation-induced electric dipole, neglecting the emission due to fluctuations. In order to do that, we just need the equation of motion of the photon operators, replacing the electric dipole operator by its mean values. The light part of the Hamiltonian becomes

$$H_L + H_{XL} = \sum_{\mathbf{q},\mathbf{m}} \left[\hbar c q b^{\dagger}_{\mathbf{q},\mathbf{m}} b_{\mathbf{q},\mathbf{m}} + \frac{g F(N_m(t))}{2} \left(e^{-i\phi_m(t)} b_{\mathbf{q},\mathbf{m}} + h.c. \right) \right]. \tag{9}$$

This term corresponds to a set of non interacting forced harmonic oscillators which can be solved for any $F(N_m(t))e^{-i\phi_m(t)}$. Assuming that the condensation starts at $t = 0$, *i.e.* that the forcing term is zero for $t < 0$, the exact single mode photonic field for $t > 0$ is [23]:

$$\begin{aligned} |\Xi_{\mathbf{q},\mathbf{m}}(t)\rangle &= e^{\Theta_{q,m}(t) - icqn_{\mathbf{q},\mathbf{m}}t} e^{iK_{q,m}(t) b^{\dagger}_{\mathbf{q},\mathbf{m}}} e^{iK^*_{q,m}(t) b_{\mathbf{q},\mathbf{m}}} |\Xi_{\mathbf{q},\mathbf{m}}(0)\rangle \\ K_{q,m}(t) &= \frac{g}{2\hbar} \int_0^t F(N_m(s)) e^{i[cqs - \phi_m(s)]} ds, \\ \Theta_{q,m}(t) &= \int_0^t K_{q,m}(s) \frac{dK^*_{q,m}(s)}{ds} ds, \end{aligned} \tag{10}$$

where $n_{\mathbf{q},\mathbf{m}} \equiv b^{\dagger}_{\mathbf{q},\mathbf{m}} b_{\mathbf{q},\mathbf{m}}$. Assuming that there are no photons before the exciton condensation, the photon field for $t > 0$ is

$$|\Xi(t)\rangle = e^{-iH_L t/\hbar} \prod_{\mathbf{q},\mathbf{m}} e^{\Theta_{q,m}(t)} e^{iK_{q,m}(t) b^{\dagger}_{\mathbf{q},\mathbf{m}}} |0\rangle \tag{11}$$

which represents a Glauber coherent state [20]. For coherent states, the expectation value of the electric field is equal to the classical electric field generated by an oscillating $\mathbf{P}_m$. All this is a consequence of

$$\langle \Xi(t) | b_{\mathbf{q},\mathbf{m}} | \Xi(t) \rangle \neq 0 \tag{12}$$

which holds even if the photon field is not in its ground state at $t = 0$. This proves our first claim that there is a coherent emission coming from an exciton condensate. Obviously, the state varies with time for the emission of one photon removes one exciton from the condensate. Also, note that Eq. (12) is not verified in the case of luminescence.

The connection between condensation and $\langle P\rangle \neq 0$ was emphasized by Sham and collaborators for an excitonic insulator. [24] As the excitonic insulator is in a true equilibrium state, the electric dipole moment associated with the condensation is time independent, in contrast to our case. A non zero $\langle P\rangle$ in an OA exciton condensate is also mentioned in reference [14], although these authors did not discuss the issue of the coherence of the emitted EMF .

We now consider the possible existence of 2D condensates at finite temperatures. Hohemberg rigorously proved that, under certain conditions, condensation is impossible in 2D systems at finite temperatures [21]. His proof relies on the divergence of the thermal depletion of the order parameter $\langle c^\dagger_{\mathbf{k}} d^\dagger_{\mathbf{k}}\rangle$ which is bigger than the integral [2,21]:

$$\int \frac{d^2\mathbf{k}}{\langle[[\rho_{\mathbf{k}}, H_X], \rho^\dagger_{\mathbf{k}}]\rangle} \tag{13}$$

where $\rho_{\mathbf{k}}$ is the $\mathbf{k}$ Fourier component of the density operator. Since H_X conmutes with the number operator, it is easy to demonstrate [25] the f-sum rule $\langle[[\rho_{\mathbf{k}}, H_X], \rho^\dagger_{\mathbf{k}}]\rangle = \hbar^2 k^2 (N_{0,1} + N_{0,-1})/2M$. If H_X is the only term in the Hamiltonian, the integral (13) contains an infrared singularity and the thermal depletion of the order parameter must be infinite, i.e., there is no condensation at finite temperature in 2D. For OA excitons, there is an additional contribution H_{XL} which must be included in the analysis. This gives rise to an extra term in the denominator of the integral (13) which suppresses the singularity. For a given value of m, the new term takes the form

$$\langle[[\rho_{\mathbf{k}}, H_{XL}], \rho^\dagger_{\mathbf{k}}]\rangle = gD_{cv} \sum_{\mathbf{q},\mathbf{p}} \langle b^\dagger_{\mathbf{q}}\rangle \left(\langle c^\dagger_{\mathbf{p}+\mathbf{k}} d^\dagger_{\mathbf{p}-\mathbf{k}}\rangle + \langle c^\dagger_{\mathbf{p}-\mathbf{k}} d^\dagger_{\mathbf{p}+\mathbf{k}}\rangle - 2\langle c^\dagger_{\mathbf{p}} d^\dagger_{\mathbf{p}}\rangle \right) + h.c. \tag{14}$$

The infrared divergency in the integral (13) disappears because Eq. (14) depends linearly on k. [4] This term is non zero due to Eq. (4) which implies Eq. (12) as shown above. In other words, long wavelength thermal fluctuations do not destroy the long range order because the condensed excitons can recombine emitting coherent light. The infrared divergency is associated with restoration of the gauge symmetry by the gapless Goldstone modes of infinite wavelength. The coupling between excitons and photons breaks this symmetry and therefore there is no gapless Goldstone modes and no divergency. The conclusion is: *optically active exciton condensation is possible in 2D at finite temperatures*

While we have established that a condensate of 2D OA excitons can exist at finite temperatures, the prediction of the critical temperature T_c is a difficult matter. Here, we give upper bounds based on our physical understanding of the problem. There are two relevant energy scales which are upper bounds for T_c [1,17]: the quantum degeneracy temperature, $T_{QD} = 2\pi\hbar^2 n/M$ below which the thermal wavelength λ_B is larger than the interparticle distance and the pairing energy, Δ, of the condensate. Numerical calculations in the BCS approximation [11] show that, for densities below $10^{11} cm^{-2}$ and $d < 100\mathring{A}$, $\Delta > T_{QD}$. Hence, exciton condensation experiments should be performed at temperatures below T_{QD}. For exciton masses of GaAs QW, T_c (in K) $\simeq 10^{-10} \times n$(in cm^{-2}) so that a typical T_c is a few degrees K.

A plausible alternative to increase the critical temperature for condensation is to use unbalanced populations $n_{+1} \neq n_{-1}$ of ± 1 excitons . To enhance the quantum degeneracy and, consequently, $T_{QD}(m) \propto N_m$ it is convenient to have all the excitons in the same state m. and, for this, one can use circularly polarized light . [19] Typically, populations tend to balance in 50 ps for a symmetric QW. This relaxation time can be increased in an asymmetric QW because the relaxation mechanism is proportional to the electron-hole overlap. Moreover, in this case a fully polarized ground state is expected [16] increasing the quantum degeneracy. In some cases, both $+1$ and -1 excitons can be condensed [16] and we expect that new physical phenomena related to the coherence between the $+1$ and the -1 phases will be uncovered [26]. However, it should be noted that the critical temperature for this two-component condensation is still lower than the one associated to the single-component condensation.

In summary, a condensate of OA excitons in a 2D heterostructure will emit coherent light even in the absence of population inversion. This effect can be used to confirm unambiguously exciton condensation. The coherent emission is an essential ingredient of condensation because it cancels long wavelength fluctuations which prevent condensation in other 2D systems.

The authors would like to thank L. Sham for a critical reading of the manuscript and D. W. Snoke for helpful comments. Work supported in part by MEC of Spain under Contract Number PB96-0085, by the Fundación Ramón Areces and by the U. S. Army Research Office under Contract Number DAAH04-96-1-0183. During the early stages of this work, R.M. was a Visiting Iberdrola Professor at the Universidad Autónoma de Madrid.

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