Four-wave mixing signatures of exciton Bose condensation

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We propose a transient grating experiment in order to observe exciton Bose condensation. We show that for typical pa in semiconductors the velocities of first and second sound are comparable, resulting in a characteristic behavior of the the longitudinal modes.

The possibility of observing Bose condensation of excitons has been a long-standing objective [1]. Since exciton parameters (scattering length, mass, etc.) are very different from those of He or spin-aligned hydrogen, and can actually be varied in various semiconductor or molecular materials, this effect could provide an important insight regarding the dynamics of quantum Bose liquids. As excitons have a finite lifetime, creating persistent superconductors is impractical even for long-lived excitons (e.g. triplet excitons or biexcitons).

Several experiments on highly excited semiconductors have been carried out in the search for Bose–Einstein condensation of excitons. Attempts focused on the predicted narrow emission lines in the luminescence spectrum of strongly pumped CuO_2 [2]. The expansion rate of an exciton gas was measured by observing the spatial distribution of the exciton gas as a function of time [3]. Later in a numerical study this was interpreted as evidence for a state with zero viscosity [4]. The interpretation of these experiments is not straightforward for several reasons: (i) The expansion rate should drop as the exciton droplet expands and the density decreases. Also there should be a transition to a normal state after some time. (ii) At the edge of the drop the density of the exciton fluid drops to zero, hence there should be a normal layer at the edge of the drop, causing friction. This may prevent the observation of a sharp transition. (iii) One should worry about whetl possible to observe superfluidity of a gas that expanding [5] – according to the Landau there is a maximum velocity difference betw normal and superfluid components. For hig locity differences the superfluidity breaks do for helium the actual breakdown velocity i lower due to the appearance of vortices. (perfluidity presupposes a hydrodynamic tion, while for a freely expanding gas a more kinetic description in terms of elementary tions (particles) seems more appropriate. M cently it has been argued that an enhanced erate four-wave mixing signal observed presence of strong pumping may suggest Bo densation [6]. It is fair to say that while all experiments may provide some evidence f densation, no unequivocal experimental opt nature has been predicted or found.

In this Letter, we analyze the use of time- nonlinear optical techniques for probing the ics of excitons at high densities. We show tha cific four-wave mixing technique, transient [7], reveals a number of characteristic feat lated to the exciton sound waves [8] whic provide unambiguous evidence for Bos densation.

We use the standard Hamiltonian

$$H=\sum_k \frac{k^2}{2m} a_k^\dagger a_k + \tfrac{1}{2}\sum_{k,p,q} V_q a_{k+q}^\dagger a_{p-q}^\dagger a_k a_p .$$

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dependent of q, a being the scattering length, and set $\hbar=1$ throughout.

As far as the equilibrium properties go, there are two dimensionless numbers characterizing the state, the density $\rho\equiv n\lambda_T^3$, with density $n=N/V$, and thermal wavelength $\lambda_T=(2\pi^2/mk_BT)^{1/2}$. The other dimensionless parameter is the interaction scattering length $x\equiv\rho a/\lambda_T$. Of these the parameter x is particularly interesting as it is large for helium but can be varied over a wide range for an exciton fluid by changing the density. It is the expansion parameter of the many perturbation theories [9,10] upon which we base our treatment. We consider a weakly interacting gas. For the normal phase the thermodynamic quantities are well known [11]. For instance for the pressure P we have

$$P=\frac{k_BT}{\lambda_T^3}g_{5/2}(z)+\frac{a}{m}n^2\,. \tag{2}$$

The second term on the right-hand side is the lowest order in density correction due to the interaction [11,12]. z is the solution to $n\lambda_T^3=g_{3/2}(z)$ with $g_\alpha(z)=\sum_n z^n/n^\alpha$. For the sound velocity we obtain

$$c_0^2=\frac{k_BT}{m}\frac{5g_{5/2}(z)}{3g_{3/2}(z)}+2n\frac{a}{m^2}\,. \tag{3}$$

For the Bose condensed phase we use the lowest-order Bogolyubov theory in which quasi-particles have energy [10,13]

$$\omega(k)=\left(\frac{aN_0}{m^2}k^2+\frac{k^4}{4m^2}\right)^{1/2}\,. \tag{4}$$

For N_0 we use a mean field theory, which is an excellent approximation as long as we are far (say a factor 2) from the critical density. Then N_0 follows as the solution of the following self-consistent equation $N=N_0+N_n$ with the number of particles in the normal phase

$$N_n=V\int\frac{d^3k}{(2\pi)^3}\frac{\omega(k)\,[\omega^2(k)+N_0^2a^2/m^2]^{-1/2}}{e^{\beta\omega(k)}-1}\,, \tag{5}$$

where we take only the thermal depletion of the ground state. When we are far away from the phase transition, N_0/N is close to unity, for a weakly interacting fluid.

culation of the thermodynamic functions thr partition sum is straightforward, we have e.g entropy

$$S=\frac{k_BV}{\lambda_T^3}\left(\tfrac{5}{2}F-x\frac{dF(x)}{dx}\right),$$

where

$$F(x)=-\frac{4}{\sqrt{\pi}}\int_0^\infty dt\,t^2\times\ln\left\{1-\exp\left[-\left(\frac{xt^2}{\pi}+t^4\right)^{1/2}\right]\right\}.$$

The ground state contribution to the pres to be taken into account separately [9]; ap the thermal contribution of quasi-particles the contribution due to wavefunction overl

$$P_0=\frac{a}{m}\left(\frac{N_0^2}{2V^2}+\frac{N_n^2}{V^2}\right).$$

The factor 2 difference between the two ter from the fact that the condensed particles a the same state. This overlap contribution c substantial difference between the sound v of the normal and superfluid phases, even m a factor $\sqrt{2}$ in a transition regime where dN large, yet N_0 is small.

To describe the time evolution of the excit sity deviation $\delta n(k,t)$ we need to consider gitudinal "normal" velocity $v_n(k,t)$, the lo nal superfluid velocity $v_s(k,t)$ and the devi the local temperature from its average $\delta T($ addition, since we do a kinetic theory we als duce additional variables, the (kinetic) stres $\sigma(k,t)$ and the temperature current $J_T(k,t$

Since the long mean free path of quasi-par a superfluid phase may destroy the observa sound waves, we model the dynamics by the ing coupled kinetic equations, which genera standard helium hydrodynamic equations [

$$\frac{d}{dt}\delta n(k,t)=ikn_nv_n(k,t)+ikn_sv_s(k,t)\,.$$

Here n_n and n_s are the normal and superflu

$$n_n = \frac{1}{6\pi^2}\int_0^\infty dk \frac{\beta k^4/m}{e^{\beta\epsilon(k)} - 2 + e^{-\beta\epsilon(k)}}, \quad (10)$$

and $n_s = n - n_n$. Furthermore

$$m\frac{d}{dt}v_n(k,t) = ik\frac{dP}{dn}\bigg|_T \frac{\delta n(k,t)}{n} + ik\left(T\frac{n_s}{nn_n}\frac{S}{V} + \frac{T}{n}\frac{dP}{dT}\bigg|_n\right)\frac{\delta T(k,t)}{T} - m\mu_v v_n(k,t) + ikm\sigma(k,t). \quad (11)$$

This equation defines $\sigma(k, t)$. For the superfluid velocity we have

$$m\frac{d}{dt}v_s(k,t) = ik\frac{dP}{dn}\bigg|_T \frac{\delta n(k,t)}{n} + ik\left(-\frac{T}{n}\frac{S}{V} + \frac{T}{n}\frac{dP}{dT}\bigg|_n\right)\frac{\delta T(k,t)}{T}, \quad (12)$$

and for the temperature deviation

$$\frac{C_V}{T}\frac{d}{dt}\delta T(k,t) = ik\left(\frac{n_s}{n}S + \frac{n_n}{n}\frac{V\,dP}{dT}\bigg|_n\right)v_n(k,t) + ik\left(-\frac{n_s}{n}S + \frac{n_s}{n}\frac{V\,dP}{dT}\bigg|_n\right)v_s(k,t) - \mu_T\frac{C_V}{T}\delta T(k,t) + ikJ_T(k,t). \quad (13)$$

This equation defines the temperature current $J_T(k, t)$. Finally for the current we have

$$\frac{d}{dt}\sigma(k,t) = ikXv_n(k,t) - \nu_\sigma\sigma(k,t), \quad (14)$$

and

$$\frac{d}{dt}J_T(k,t) = ikY\delta T(k,t) - \nu_J J_T(k,t). \quad (15)$$

The terms proportional to the μ describe the effect of the acoustic (lattice) phonons, the terms proportional to ν are caused by quasi-particle–quasi-particle scattering. To lowest-order approximation $\nu \propto \nu_0 \equiv n_n \pi a^2 \bar{v}$, where a is the scattering length, and $\bar{v}$ is the average velocity which can be estimated to

$$\bar{v}^2 = \frac{1}{6\pi^2 m^3}\int_0^\infty dk \frac{\beta k^6}{e^{\beta\epsilon(k)} - 2 + e^{-\beta\epsilon(k)}}.$$

The exciton–exciton interaction introduce parameter, the wavevector

$$k_0 \equiv na^2.$$

It should be pointed out that N_n/N is not n_n/n, as the latter is the "dynamic" normal as defined by Landau [9]. We use the Landa throughout. We have taken the classical flui $X = \frac{4}{5}Y = \frac{4}{3}\bar{v}^2$. For a classical fluid $\bar{v}^2 = k_B T/$

For a typical experimental condition in C have $m = 3m_{el}$ [14], $n = 10^{19}$ cm^{-3} [2]. As pulse that creates the excitons also heats up tice the temperature of the exciton gas may high. For the scattering length we take two ti exciton Bohr radius, $a = 1.4$ nm. If we take for the temperature we obtain $\rho = 20$ and $x=$ are in an intermediate regime, quite differe superfluid helium.

The proposed transient grating experime with a strong laser pulse that creates the excit We then apply a pair of time-coincident pulses with wavevectors $\boldsymbol{k}_1$, and $\boldsymbol{k}_2$ that set exciton grating with wavevector $\boldsymbol{k}_G = \boldsymbol{k}_1 - \boldsymbol{k}_2$. fixed time delay, a weak off resonant probe pu wavevector $\boldsymbol{k}_3$ is applied that is diffracted remnant of the grating of the first pulse to a signal with wavevector $\boldsymbol{k}_s = \boldsymbol{k}_3 + \boldsymbol{k}_G$. The i of the diffracted signal is proportio $S(t) = |\delta n(\boldsymbol{k}_G, t)|^2$.

We solve eqs. (9)–(15) with the initial co $\delta n(\boldsymbol{k}, t=0) = 1$, and the other variables ze signal amplitude $\delta n(\boldsymbol{k}_G, t)$ can then be expa the six modes

$$\delta n(\boldsymbol{k}_G, t) = \sum_{j=1}^{6} c_j \exp(iv_j k_G t - \Gamma_j k_G^2 t).$$

There are four eigenmodes with eige $\pm iv_j(k) - \Gamma_j(k)k^2$ corresponding to first soun 2) and second sound ($j = 3, 4$), and two modes with $v_j = 0$. Γ_j is the damping (tra coefficient. In the following calculations we exciton–phonon interactions so $\mu_{v,T} = 0$.

In fig. 1 we present the velocities of first

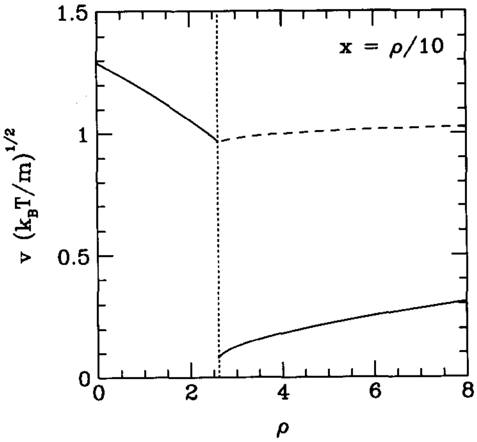

Fig. 1. The sound velocity as a function of density. The dotted line indicates the transition density. The two lines correspond to first sound, a density mode (solid) and second sound, a temperature mode (dashed). This identification follows from a study of the eigenfunctions. At these densities only first sound can be observed in a transient grating experiment. This figure suggests that the velocity of density oscillations is discontinuous at the superfluid transition and that second sound, not first sound, is to be considered as the extension of normal sound.

continuity of the sound velocity at the transition may be due to our use of a mean field description and may be rounded off in reality. We notice a large decrease in the velocity of first sound when we cross the transition density. While there is a sound mode with roughly the same velocity in the normal and superfluid phases, it does not correspond to density oscillations in the superfluid phase and hence cannot be observed in a transient grating experiment. The drop in the sound velocity is the first signature of Bose condensation that can be obtained from a transient grating experiment.

The two sound modes for higher density are presented in fig. 2. For a highly degenerate fluid (x large) such as helium we have the well-known expressions for the sound velocities. For first sound

$$v_1^2 = \frac{n_s}{mn} \left. \frac{dP}{dn} \right|_T , \tag{19}$$

and for second sound

$$v_2^2 = \frac{V}{mC_V} \left(T \frac{n_s}{nn_n} \frac{S}{V} + \frac{T}{n} \left. \frac{dP}{dT} \right|_n \right)^2 . \tag{20}$$

In the coupled equations we find a curve crossing.

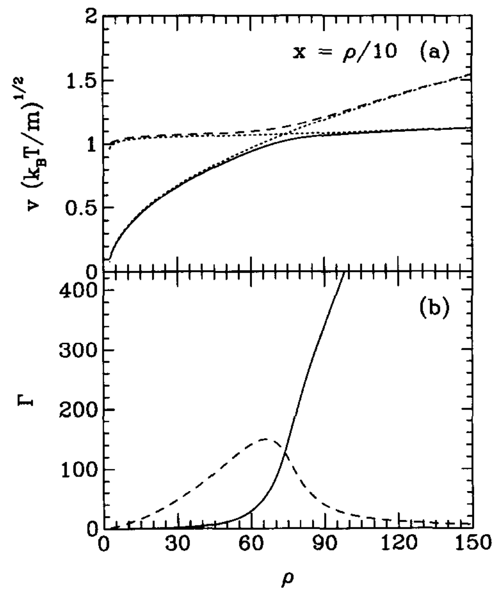

Fig. 2. (a) The velocity of the hydrodynamic modes v_j as a function of density for much higher density than presented in fig. 1. Dotted: lowest-order approximation eqs. (19) and (20). Dashed and solid: two of the eigenvalues obtained by diagonalizing the 6×6 matrix eqs. (9)–(15). (b) The decay rate Γ_j of the two modes displayed in (a) in the hydrodynamic regime as function of density. The decay is in units $(k_BT/m)^{1/2}/k_0$. In addition there are two nonpropagating modes (not shown).

The mode with the lower damping turns out to be the density mode that shows up in the grating. There is a density range near the crossing ($\rho = 70$ in fig. 2) where the damping of both modes is comparable.

The physical origin is the following. For a weakly interacting fluid x is very small. Then dP/dn is very small, for an ideal Bose fluid it is actually zero, so that density waves (first sound) is nonpropagating. On the other hand, for a strongly interacting fluid first sound is faster than second sound by a factor $\sqrt{3}$, and for a certain parameter the velocities of first and second sound become comparable. At this state point density oscillations are damped. This is the second signature of Bose condensation.

The temporal profile of the grating response is shown in fig. 3 for three state points. The times at which the grating vanishes produces a direct measurement of the sound velocity. In figs. 3a and 3b the grating response has an oscillating envelope, demonstrating the presence of two sound modes. In fig. 3c the mean free length is too large to observe second

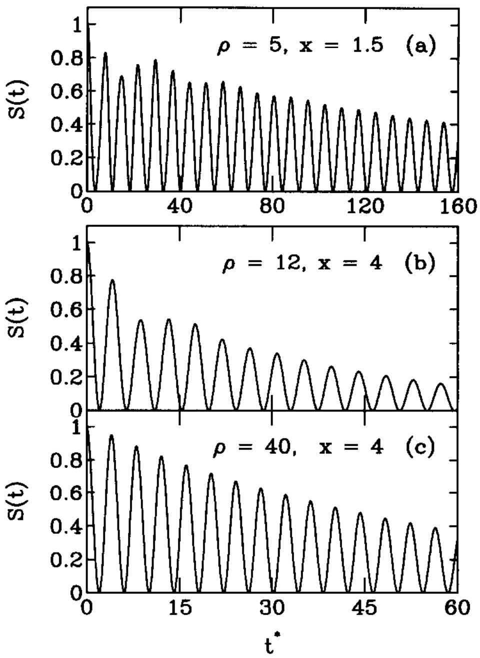

Fig. 3. The grating signal for suitable phase points and wavevector $k_G/k_0=0.005$. The state point in (a) corresponds to $n=6\times10^{19}$ cm^{-3}, and $T=85$ K. For these values the wavevector corresponds to $k\approx2(\mu m)^{-1}$. The time is in units $k(k_BT/m)^{-1/2}$ corresponding to about 60 ps. The beats in (a) and (b) reflect the coexistence of two sound waves. In (c) the mean free length is too large for second sound to exist. For a normal fluid the grating decays like in (c).

sound. These results suggest a third experimental probe of Bose condensation. We have thus three characteristic signatures of Bose condensation in a transient grating experiment.

The light mass of excitons (close to the electron mass) makes it easier to observe Bose condensation compared with atomic systems (spin-aligned hydrogen or He). However, their finite lifetime is a serious limitation. Certainly excitons have to be sufficiently long lived to allow for the transition. Optically forbidden triplet excitons or biexcitons [6], which are longer lived, are therefore preferable. Exciton annihilation processes which are faster at high exciton densities pose an additional difficulty. Nevertheless, densities of 10^{19} cm^{-3} were achieved in CuCl without this becoming a problem [2]. For anthracene densities of the order of 10^{23} cm^{-3} are needed to observe exciton fusion [15]. The formation of the condensate has attracted considerable theoretical attention [16]. For a system not too far from equilibrium the exchange between normal and condensed particles is very rapid. This is based on the observation that the effective Hamiltonian is not number conserving, and using the Bogolyubov eigenfunctions it can easily be shown that there is an oscillation between quasi-particles with momentum q and $-q$ that has a frequency ϵ_q. While this exchange is collisionless, due to the distribution of frequencies there is a dephasing and we expect the exchange to take place on the timescale of $t_x=\hbar/k_BT$, which is of the order of picoseconds. One could therefore observe Bose condensation even if the particles have a finite lifetime.

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