Exciton Transport in Cu_2O: Evidence for Excitonic Superfluidity?

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(Received 15 March 1993)

A marked reduction in the velocity dispersion of excitons propagating through a thick Cu_2O crystal at low temperatures and high densities is attributed to the onset of a superfluid excitonic phase.

PACS numbers: 71.35.+z

Superfluidity in liquid ^{4}He is intimately connected with Bose-Einstein condensation. However, because of the high density of atoms in the liquid, it is not easy to disentangle the effects due to particle-particle interactions (required for superfluidity) from the pure quantum statistical effects leading to Bose-Einstein condensation (BEC) [1]. For instance, it is known that below 1 K, the total mass of the liquid is superfluid yet only about 10% of the particles are found to be in the condensed state. It is therefore important to find other, more dilute systems where the connection between Bose statistics and superfluidity can be studied as a function of particle density.

It has been recognized in the past decade that excitonic particles in semiconductors can display the quantum statistical properties expected from bosons [2]. In particular, it has been shown recently that exciton densities exceeding the critical density for BEC can be obtained in Cu_2O [3]. An important question concerns exciton transport properties under such conditions. Does the excitonic gas become superfluid, a regime characterized by a drag-free motion of part of the fluid? For excitons propagating freely inside a crystal, this corresponds to the disappearance of scattering of excitons with acoustic phonons, a process which represents the main source of drag in their motion. The study of superfluid motion and its relation with BEC is particularly interesting in an excitonic fluid since the particle density necessary for BEC can be much lower than in liquid He, due to the very small mass of excitonic particles, thereby minimizing the effects from interactions. For example, in the case of excitons in Cu_2O with a Bohr radius $a_B = 0.7$ nm, the critical density n_c for Bose condensation is 10^{17} cm^{-3} at $T = 2$ K so that $n_c a_B^3 \ll 1$ and one truly deals with a weakly interacting Bose gas.

There are some indications that a drastic reduction of friction in the motion of excitons takes place at high density in Cu_2O. Anomalies of transport have been observed through time and spatially resolved measurements of the exciton luminescence at early times following particle creation [3]. At high densities, the front of the excitonic cloud expands ballistically at near sonic velocity away from the surface where it is initially located. By contrast, similar measurements at low densities reveal a normal diffusive regime [4]. It has been proposed in Ref. [3] that this behavior signals the onset of superfluidity. Recently, Link and Baym [5] have come to a similar conclusion from a numerical simulation of the hydrodynamics of the excitonic gas at early times. They concluded that the rapid initial expansion of the exciton gas indicates a substantial reduction of drag, supporting an interpretation in terms of superfluidity. However, another interpretation of the results based on a classical model has been proposed by Bulatov and Tikhodeev [6]. In this alternate explanation, the exciton gas is normal, i.e., noncondensed, but it is pushed towards the interior of the sample by a phonon wind emanating from the surface.

In this Letter, we investigate the transport properties of an exciton gas in Cu_2O at much later times. It is then easier to distinguish between diffusive and superfluid behavior since diffusion leads to a characteristic spatial spread of particles which increases with the elapsed time. For a local, time-resolved detection of the particles several mm away from the initial excitation spot, we have recorded the photovoltaic current resulting from exciton dissociation at a semiconductor-metal interface [7]. This method of detection is appropriate for long-lived paraexcitons in Cu_2O [8], which have a weak radiative recombination rate and are therefore difficult to observe by luminescence. The results bring clear evidence for the onset of an anomalous particle current at sufficiently high particle density and below a critical temperature. The temperature and density dependence of this new transport regime is difficult to explain by a diffusion model even in the presence of a phonon wind but is consistent with an interpretation in terms of superfluidity of an excitonic condensate.

In the experiments, the front surface of a high quality, natural-growth Cu^2O single crystal in the form of a parallelepiped of dimensions as shown in Fig. 1 is irradiated with light pulses of 10 ns duration at a wavelength $\lambda = 532$ nm obtained by second harmonic generation of the output from a Q-switched Nd:YAG laser. The incident beam 2 mm in diameter corresponds to the central region of a parallel beam with 1 cm diameter, thereby

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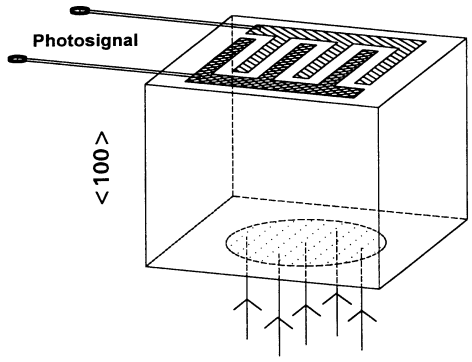

FIG. 1. Schematic representation of the sample with the electrodes to detect the photocurrent. The fingers consisting of evaporated gold and copper are 200 μm wide and separated by a distance of 200 μm. Arrows represent the incoming laser beam. The sample is a natural growth single crystal with faces oriented along the (1,0,0), (0,1,0), and (0,0,1) crystal axis. All dimensions are in rough relative scale.

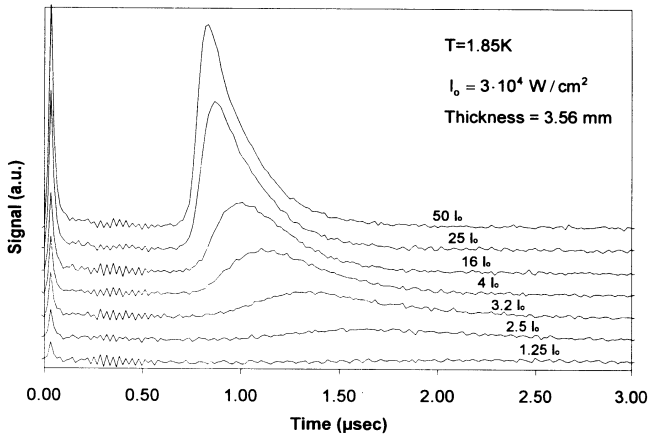

FIG. 2. Time-resolved photovoltaic voltage measured across an external 50 Ω resistance for different incident laser intensities. The fast signal at $t=0$ results from direct illumination of the surface containing the electrodes and gives the detector response time. The sample of thickness 3.56 mm is immersed in liquid He at $T=1.85$ K.

ensuring good uniform illumination of the front surface of the sample. The incident green light of maximum intensity 10^7 W/cm^2 is absorbed over a distance of the order of $\alpha^{-1} \approx 1$ μm, where α is the crystal absorption coefficient at $\lambda = 532$ nm, resulting in the formation of a high density of excitons in the form of a thin pancake located initially in the proximity of the front sample surface. On the rear side of the sample a comb structure of copper and gold finger contacts as shown in Fig. 1 acts as a local detector of excitons [7]. Particles reaching the immediate vicinity of the Cu/Cu_2O interface dissociate into free carriers under the effect of the strong built-in electric field. The liberated holes are swept across the contact barrier while the free electrons cross the Ohmic gold contacts, resulting in an external electric current proportional to the local exciton density which we measure across a 50 Ω load resistance.

Results are shown in Fig. 2 for a crystal of thickness $d = 3.56$ mm immersed in liquid helium at a temperature of 1.85 K for different incident intensities. One can distinguish two signals. The first prompt photovoltaic response is due to intentional direct illumination of the rear surface and provides a fiducial point for time $t=0$. (This prompt signal is removed by shielding the sample from incident light except for the illuminated spot at the front sample surface.) It also indicates the response time of the exciton detector. The delayed photovoltaic response occurring about 1 μs later is due to excitons having propagated through the sample. It can be clearly seen that the exciton transport undergoes a qualitative change over a small range of particle densities. The response obtained at the lowest exciton densities shows a large spread of exciton transit times which is typical of the diffusive behavior from a free gas expanding through a viscous medium. However, at higher densities a sharp decrease by nearly 2 orders of magnitude in the spread of transit times occurs; the majority of particles now propagate together at a common speed v nearly equal to but below the longitudinal sound velocity in the medium $v_l = 4.5 \times 10^5$ cm/s [9] independent of the crystal thickness. Measurements performed with a thicker sample ($d = 8.85$ mm) at the same temperature confirm a drag-free motion of a packet at similar velocity $v \leq v_l$ provided the incident intensity exceeds 0.1 MW/cm^2 as shown in Fig. 3. Figure 4 shows the photovoltaic signal as a function of temperature at constant input light intensity. Here again propagation of the exciton packet is in the form of a ballistic exciton packet if $T < 3$ K but becomes diffusive at higher T. Note the very small temperature

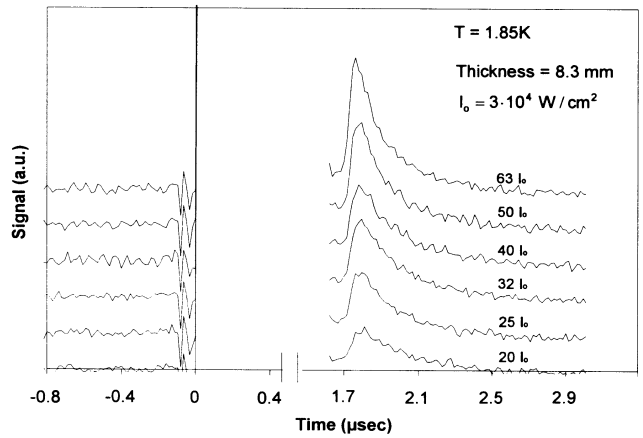

FIG. 3. Time-resolved photovoltaic voltage measured across an external 50 Ω resistance for different incident laser intensities in a sample of thickness 8.3 mm at $T=1.85$ K.

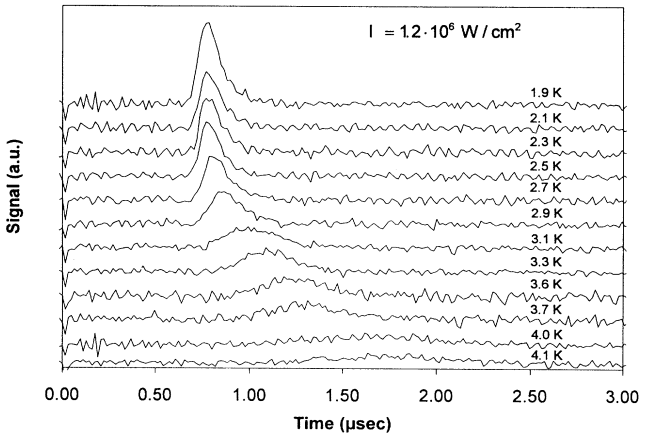

FIG. 4. Time-resolved photovoltaic voltage in a 3.56 mm thick sample as a function of sample temperature for a fixed incident laser intensity 1.2×10^6 W/cm^2.

interval over which this transition takes place.

Does this behavior reflect excitonic superfluidity? To bring an element of answer to this question we first compare the density of particles inside the packet to the critical density for Bose-Einstein condensation of excitons as predicted by the ideal Bose model. Since the measurements are performed at long delays, it is expected that all particles of the initial mixture of ortho- and paraexcitons have converted into energetically lower paraexcitons of long lifetime $\tau > 10$ μs [8]. Also, the temperature of the excitonic fluid is now imposed by the crystal temperature, in contrast to the situation at early times $t \approx 10^{-9}$ s where excitons can acquire an effective temperature significantly higher than that of the lattice. The density after 1 μs can be estimated from a knowledge of the total number of particles created initially and from the volume of the packet, as given by its thickness $d \approx \Delta t v_l$, where Δt is the temporal width (FWHM) of the packet and taking the lateral dimensions as equal to the initial ones. Assuming negligible decay of long-lived paraexcitons until their detection, we find for an excitation level $I = 20 I_0 = 600$ kW/cm^2 (corresponding to the experimental conditions for the appearance of the ballistic excitonic packet in Fig. 2) a value n ($t = 1$ μs) $\approx 1.2\times10^{17}$ cm^{-3} where a factor of 2 has been included to take into account losses at the front surface [10]. This is in good agreement with the calculated critical density $n_c = 6\times10^{16}$ cm^{-3} for paraexcitons of mass $m = 2.7 m_0$ at $T = 1.85$ K [11]. On the other hand, for a density of $n \approx 2.5\times10^{17}$ cm^{-3} corresponding to the case of Fig. 4 the critical temperature is $T \approx 3$ K so that a superfluid packet linked to the onset of BEC should disappear above this temperature, again in agreement with the results of Fig. 4. In view of the sharpness of the transition from ballistic to diffusive motion, an explanation in terms of a phonon wind pushing classical particles is unlikely. On the other hand, in view of the close agreement with the predictions of the ideal Bose gas, it is plausible to associate the new propagation mode to the onset of Bose condensation.

Why does the superfluid occur in the form of a particle packet propagating at nearly the sound velocity? We believe this reflects the rather unique initial conditions of the fluid. As shown in Ref. [3], the initial nonequilibrium conditions lead to a local supercritical paraexciton density despite the important increase of excitonic effective temperature. Such a strongly localized condensate is subject to the combination of several effects. On the one hand, the steep gradient in the chemical potential between excited and nonexcited regions of the crystal acts as a force pushing the superfluid. One expects therefore a particle current flowing without drag towards the interior of the crystal. On the other hand, once established, a condensate is not easily destroyed since it is a collective effect, with all particles in the condensate mutually attracting each other. Because of the finite duration of the excitation source, the moving condensate must then take the form of a packet of limited size in order to conserve the total number of condensed particles. We note that in the absence of drag such a packet can be easily accelerated by any force (including a phonon wind), but only up to a maximum velocity given by the longitudinal sound velocity of the medium [12]. For higher velocities the superfluid becomes unstable, since energy dissipation can take place by emission of acoustic phonons and simultaneous removal of individual particles from the condensate. As a net result one expects a stable superfluid to propagate ballistically at near sonic velocity in the form of a localized particle packet.

A quantitative treatment of the problem is a challenging task [13]. Among the questions which need clarification many pertain to the dynamics of Bose condensation and superfluid formation. How long does it take for the condensate and the superfluid to be formed? What determines the speed of the packet? Is the ballistic speed fixed by early conditions when the density of particles and therefore the interaction potential between particles are higher? Answers to these questions require solving the Gross-Pitaevskii [14] nonlinear equation,

$$ih\partial\phi/\partial t = -h^2/2m\Delta\phi + g|\phi|^2\phi, \qquad (1)$$

under the appropriate initial conditions, where $\phi(r,t) = n_s^{1/2}\exp -i\varphi(r,t)$ is a classical wave function of large amplitude describing the condensate with $\varphi(r,t)$ its phase factor, n_s its density, and $g = 4\pi h^2 f/m$ with f is the amplitude of exciton-exciton scattering.

In this context, it is worth noting that a family of solutions to the nonlinear Schrödinger equation (1) is given by solitons as shown by Zakharov and Shabat [15]. The solitons determine the asymptotic behavior of large amplitude wave functions as $t \rightarrow \infty$. For a repulsive potential ($g > 0$) dark solitons (i.e., a traveling void of particles in a spatially uniform condensate) are obtained as

solutions to Eq. (1). On the other hand, bright solitons (solitary crest) require an attractive potential $g < 0$. According to Keldysh and Koslov [16], excitonic superfluidity is obtained in a fluid composed of excitons with equal electron and hole effective masses m_e and m_h (a case applying well to Cu_2O with $m_e = 0.7 m_h$) because the weak attractive potential between excitons at large distances is counterbalanced at short distance by the repulsive potential reflecting the Pauli exclusion principle acting on the Fermi particles constituting the excitons. It would be interesting to explore whether such a superfluid could also lead to bright-soliton solutions [17].

In conclusion we have reported an unusual propagation regime for excitons in Cu_2O. Above a critical particle density and below a critical temperature the particles propagate together ballistically at nearly the speed of sound of the medium in the form of a quasistable packet over unusually large distances, of the order of 1 cm. The estimated particle density and temperature required for this effect to occur agree with the conditions required for a gas of bosons to undergo Bose-Einstein condensation. We believe this behavior reflects the motion of an excitonic superfluid which is initially strongly confined spatially and evolves at later times in the form of a quasistable wave packet which shows little spatial dispersion.

We acknowledge a fruitful discussion with Dr. A. Ivanov and the assistance of E. Benson and G. Lafrenière during the measurements.

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