Progress in the Bose-Einstein Condensation of Biexcitons in CuCl

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The occupancy of the $K=0$ ground state of the biexciton in CuCl is measured by nonlinear optical methods. An increase of the optical phase conjugation signal is observed when a random high density of biexcitons is injected into the crystal. The corresponding increase of coherency is attributed to a quantum attraction towards a Bose-Einstein condensate of biexcitons.

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The recent spectacular developments in the cooling of atoms raise the exciting prospect that Bose-Einstein condensation (BEC) in a dilute atomic gas will be soon achieved. Indeed, densities of hydrogen atoms as high as $n=8\times10^{13}$ cm^{-3} have been recently reported at a temperature $T\cong100$ μK, only a factor of 3.5 above the critical temperature of this phase transition [1].

Excitonic particles in semiconductors offer an interesting alternate system [2–4] for the observation of Bose-Einstein condensation. Excitons have the advantage of a light effective mass, typically of the order or less than the free electron mass, resulting in comfortably higher critical temperatures for this phase transition. Also, since excitonic particles can be created or destroyed by absorption or emission of photons, several properties of BEC can be conveniently studied by optical methods. For instance, a straightforward approach consists in performing a spectral analysis of the luminescence due to the radiative decay of excitonic particles. Evidence for BEC is sought in the appearance, above a threshold particle density, of a sharp emission line. This line signaling the accumulation of a large number of particles in the lowest-energy state $K=0$ should be accompanied by an emission tail due to excited particles coexisting with the condensate. In the weak interaction limit, this tail should have a characteristic spectral shape consistent with a Bose-Einstein distribution function with a zero value of the associated chemical potential μ.

Bose-Einstein quantum statistics have been verified in a gas of free excitons in Cu_2O over the range covering the classical limit $|\mu|\gg kT$ up to the saturation limit $\mu\cong0$. Results are in excellent agreement with the ideal Bose gas model [5,6]. Recently, densities of paraexcitons exceeding the critical density have been reported in the same material, accompanied by anomalies in the emission spectrum and transport properties which suggest the onset of excitonic superfluidity [7].

However, Cu_2O represents a special case for two reasons. First, excitons in Cu_2O do not form biexcitons at high densities [8]. In most other semiconductors, Bose-Einstein condensation is expected to occur inside the molecular rather than the atomic form of the excitonic fluid. Second, excitons in Cu_2O do not couple to the radiation field in the dipole approximation. In materials with dipole-allowed interband transitions the direct observation by luminescence of a condensate of biexcitons at $K=0$ is prevented by the strong dispersion near $K=0$ of the polariton which is the final state in the biexciton radiative decay [4]. On the other hand, the observation of a Bose condensate of biexcitons at a finite K value is possible by luminescence and has been reported in CuCl [9,10]. Although many of the features observed in this case are those expected from a Bose condensation of biexcitons [10], this evidence has remained controversial, because of the coherent laser source used for creating biexcitons directly by resonant two-photon absorption.

In this Letter we report on a new experimental approach to the study of BEC of $K=0$ biexcitons. This method, which applies nonlinear optical methods to the detection of BEC of biexcitons, has the merit that it directly addresses an essential attribute of a Bose condensate, ignored in luminescence detection schemes, namely, its coherency. As will be discussed below, the temporal and spatial coherency resulting from the macroscopic occupation of the $K=0$ biexciton state can be probed by monitoring an increase of the phase conjugate signal of a crystal in which a Bose condensate of biexcitons is present. An optical phase conjugate signal beam is emitted from a crystal if three incident optical beams, consisting of two counterpropagating pump beams of frequency ω and a noncollinear probe beam of the same frequency, interact via the nonlinear optical response of the medium [11]. Usually, the generation of the signal beam corresponds to the scattering of one of the pump beams from the grating formed by the nonlinear interference of the other pump beam and the probe beam (real-time holo-

graphic analog). However, in the case of a nonlinear response dominated by a two-photon resonance $E=2\hbar\omega$ (as is the case here), the signal beam corresponds to the scattering of the probe beam from a spatially uniform two-photon coherence induced by the two incident pump beams. In our experiment, macroscopic coherent excitation of the biexciton two-photon resonance is achieved through injection of a high density of free excitons followed by random biexciton formation and condensation. Therefore the observed increase of coherency results from a true self-organization of the system into a state of low entropy once the critical biexciton density is reached.

Thin single-crystal platelets of CuCl, typically 10 μm in thickness, were prepared by vapor phase deposition. High-quality samples were selected from their excitonic luminescence at low temperature under weak optical excitation. An excimer laser was used to pump the dye lasers (BBQ in p-dioxane) providing the three beams required to obtain a phase conjugate (PC) signal. The two counterpropagating pump beams had opposite circular polarization vectors (in the crystal reference frame). The pump beams were focused on the sample surface to a spot of 100 μm diameter, resulting in a total intensity of 100 kW/cm^2 for a pulse duration of 10 ns. The weaker probe beam ($I_0\cong 15$ kW/cm^2), also circularly polarized, was focused on the central part of the sample region irradiated by the pump beams. The external angle between pump and probe propagation axis was 10 deg, corresponding to an internal angle of 3.6 deg. The laser linewidth was typically 0.026 meV (FWHM). The PC signal was detected around the peak of the biexciton resonance through an aperture of 2 mm diameter at a distance of 3 m from the sample immersed in superfluid He. Its intensity was monitored at $\hbar\omega=E_{xx}/2=3.1858$ eV [12,13] which corresponds to the peak of the biexciton resonance in the degenerate regime or at $\hbar\omega=3.1855$ eV in the nondegenerate regime in which case the photon energy of the probe beam was set at 3.1862 eV. It was verified that the PC signal disappeared if any of the pump or probe beams was blocked and that the signal had a circular polarization opposite to that of the probe beam.

In order to create a high density of biexcitons above the critical density for BEC, we used the broadband superradiant emission from a dye cell pumped with the same excimer laser. The intensity of the incoherent pump source could be continuously adjusted with a variable intensity attenuator up to a maximum intensity of 440 kW/cm^2. The spectral width of the superradiant emission was limited to 16 meV with a filtering monochromator adjusted to the energy of the Z_3 exciton absorption around $E=3.204$ eV. In some experiments, a monochromatic dye laser beam was used instead of the broadband source. Since in each case biexcitons are formed via exciton injection followed by molecular formation through exciton-exciton collisions, the supply of biexcitons is always random, without memory from the coherency of the initial pump source. We therefore indifferently refer in the following to these sources as the incoherent pump source.

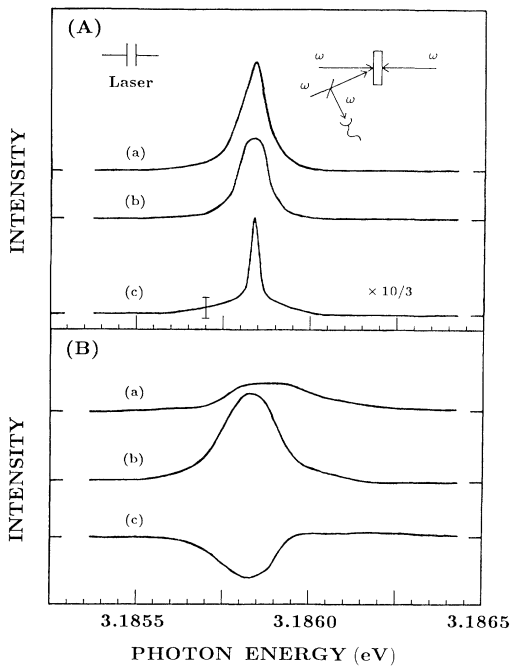

FIG. 1. Photon energy dependence of the phase conjugate signal at 2 K for two CuCl samples of different quality shown successively (curves a) in the presence of and (curves b) without incoherent pump. Curves c, the differential signal $a-b$. Sample (A) of highest quality has a thickness of 21 μm; sample (B) a thickness of 7 μm. The intensities of the counterpropagating pump, probe, and incoherent beams are 50 kW/cm^2, 50 kW/cm^2, 15 kW/cm^2, and 400 kW/cm^2, respectively.

Figure 1 shows the PC signal as a function of the laser photon energy, recorded in two representative samples in the presence (curves a) and the absence (curves b) of the incoherent pump source, and the corresponding difference signal (curves c). In the sample of highest quality (type 1 samples), displaying a very narrow PC spectrum, we note that under incoherent pumping the conjugate signal intensity is enhanced by as much as 30% (Fig. 1, curve c, top panel). We also note that this enhancement of the PC signal occurs at the peak of the biexciton resonance over a very narrow spectral width of 25 μeV corresponding to the probe beam bandwidth. In the nondegenerate regime a similar enhancement of 45% is observed in other samples of similar quality and thickness. In type-2 samples of lesser quality for which a slightly broader PC spectrum and weaker biexciton luminescence efficiency is observed, the differential PC signal is negative as shown in the bottom panel of Fig. 1. In Fig. 2, we show the enhancement of the PC signal as a function of the incoherent pump wavelength. Clearly, an increase of the conjugate signal is only observed for high injected exciton densities, i.e., in the region of strong linear absorption

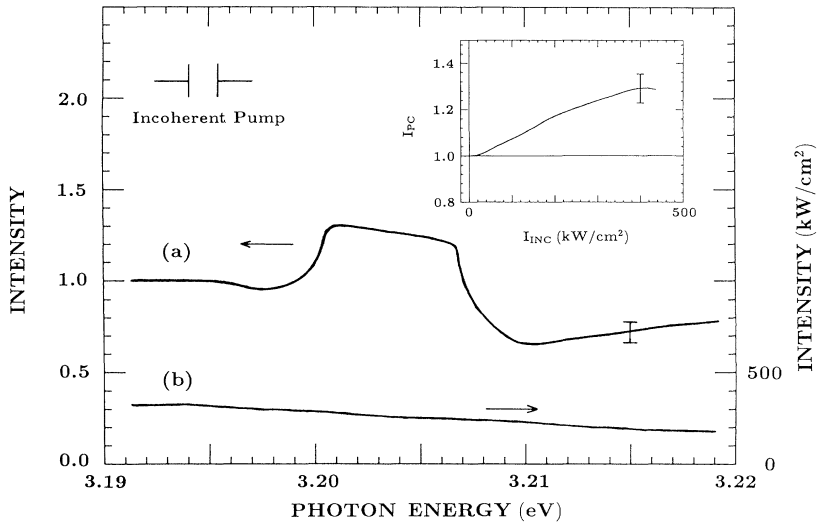

FIG. 2. Curve *a*, variation of the PC signal with narrow band incoherent pump photon energy. Curve *b*, the intensity of the incoherent pump. In the inset is shown the dependence of PC signal with incoherent pump intensity. Other parameters are the same as in Fig. 1(A).

around 3.204 eV due to the Z_3 exciton, whereas the reversed trend is observed in the region 3.21 eV corresponding to weaker absorption and lower particle density. In the inset we show the dependence of the PC signal with broadband incoherent pump intensity.

To interpret these surprising results, it is instructive to examine the time ordering of the photon events responsible for the PC signal, first in the absence of incoherent pumping. At the peak of the biexciton resonance, $2\hbar\omega = E_{xx} = 6.372$ eV, the transition is dominated by the sequence shown in Fig. 3. In each elementary step, two photons, one from each laser pump beam, are absorbed, leading to a coherent oscillation at frequency 2ω which is spatially uniform over the excited crystal volume. In a particle picture, a population of biexcitons in the $K=0$ state is created uniformly throughout the sample volume. The probe beam then induces a two-photon transition back to the crystal ground state, with the signal photon emitted in the direction opposite to the incoming probe photon. As is well known [11], the conjugate signal beam possesses time reversal symmetry with respect to the probe beam. This provides a sensitive means of selecting a very narrow region of phase space for the emitting biexcitons, simply by spatial filtering of the signal beam. For instance, in our case the internal solid angle of detection was 4.6×10^{-6} sr.

Now it is well documented in semiconductors that collision processes lead to a redistribution of excitonic particles in phase space. This has been demonstrated, for instance, in photon echo experiments, where the injection of neutral or charged particles leads to a drastic reduction of the dephasing time of a prepared coherent excitonic state [14]. In our context, the presence of a high density of excitons, from the incoherent pump source, should lead to a reduction of the conjugate signal because of collisions with the biexcitons at $K=0$ as observed in type-2 samples. On the other hand, the observed opposite trend seen in type-1 samples can be simply understood by considering the quantum behavior of Bose particles. Once the critical value for Bose condensation N_c is reached, all supplementary particles collect in the single quantum state $K=0$. This increased population in the lowest quantum state will reinforce the amplitude of the spatially uniform coherent oscillation at frequency 2ω and therefore the magnitude of the conjugate signal.

In order to check whether the density of biexcitons created by the incoherent pump does indeed exceed N_c, we compared the corresponding biexciton luminescence intensity to that obtained with a single linearly polarized laser beam tuned to the biexciton two-photon absorption peak. The density of biexcitons from the laser source can be estimated by measuring the two-photon absorbance of the transmitted laser beam. We obtain a density of

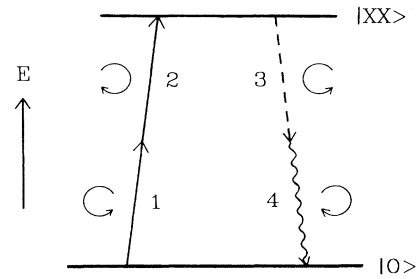

FIG. 3. Schematic diagram showing the sequence of photon events leading to the PC signal at the biexciton resonance. 1 and 2 are the counterpropagating pump photons; 3 and 4 the probe and signal photons.

4.7×10^{14} cm^{-3} biexcitons for a laser intensity $I_p=100$ kW/cm^2 assuming a biexciton lifetime of 100 ps [15]. The total biexciton population from the incoherent source can be obtained by multiplying this value by Cd/d_l where $C=41$ is the ratio of biexciton luminescence intensity due to the incoherent pump ($I=400$ kW/cm^2) and coherent pump ($I_c=100$ kW/cm^2), $d_l<1$ μm is the effective crystal thickness pumped by the incoherent beam, and $d=21$ μm is the total crystal thickness. Finally, we obtain $N>4.1\times10^{17}$ cm^{-3} for our experimental conditions. For a gas of biexcitons in CuCl with mass $m_{xx}=5.2m_0$ [16], $N_c=2\times10^{17}$ cm^{-3} at 2 K, showing that the critical density for Bose condensation is indeed obtained by the use of our incoherent pump.

It is interesting to estimate the occupation number per biexciton mode around $K=0$ corresponding to the change of the conjugate signal of Fig. 1. The total number of radiating biexciton modes is fixed by the excited sample volume, the spectral resolution of the detection, and the detection solid angle. Taking all these factors into account, we estimate a maximum of less than 22 modes seen by the detector. Assuming that the phase conjugate signal is equally distributed among these modes, we find that the occupation number of the $K=0$ biexciton state is at least 1.1×10^6.

Before concluding, we wish to make a final comment about the kinetics of BEC formation. As noted above, the biexciton density from the incoherent pump source alone exceeds the critical density. Therefore, if the time for BEC formation is shorter than the incoherent pump duration, one should expect the appearance of a conjugate signal even in the absence of the counterpropagating pump sources. So far we were unable to detect any conjugate signal under such conditions within the noise limit. The time required for the formation of a Bose quasicondensate has been recently calculated by several groups [17–20]. Kagan, Svistunov, and Shlyapnikov [20] distinguish two regimes: formation of a quasicondensate characterized by the suppression of density fluctuations which is fast, and formation of a true condensate with suppressed phase fluctuations and long-range order, which proceeds at a slower rate. On the other hand, the initial presence of a coherent signal at $K=0$ may act as a seed precipitating the phase transition into a true condensate [17]. Our results, which are sensitive to the phase coherence, seem to indicate that such a seed (provided by the two coherent counterpropagating pump sources) is necessary to obtain a true condensate within the pump duration.

In conclusion, we interpret an observed increase of the optical conjugate signal as evidence that a Bose condensation of biexcitons takes place in high-quality CuCl crystals. The required density of biexcitons is obtained through bimolecular formation from a random source of excitons. The presence of a weak population of biexcitons directly pumped into state $K=0$ seems to play an important role in the nucleation of this phase transition.

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