Trapping Long-Lifetime Excitons in a Two-Dimensional Harmonic Potential

D.W. Snoke,* Y. Liu, and Z. Vörös

Department of Physics and Astronomy, University of Pittsburgh,
3941 O’Hara St., Pittsburgh, PA 15260

L. Pfeiffer and K. West

Bell Labs, Lucent Technologies, 700 Mountain Ave., Murray Hill, NJ 07974

Abstract

We report an important step forward for the goal of unambiguous observation of Bose-Einstein condensation of excitons in semiconductors. We have demonstrated a system in which excitons live for microseconds, much longer than their thermalization time, move over distances of hundreds of microns, and can be trapped in a harmonic potential exactly analogous to the traps for atomic condensates. We also report recent results of a new method for observing evidence of Bose-Einstein condensation, by angular resolution of the emitted luminescence.

*Electronic address: snoke@pitt.edu
As far back as the 1960’s, theorists have predicted[1, 2, 3, 4, 5, 6, 7] that excitons in semiconductors should undergo Bose-Einstein condensation (BEC) at low temperature. The initial attempts at observing this effect used bulk semiconductor crystals[8, 9, 10], but gave ambiguous results. Some of the barriers to observation of this effect include a competing phase transition at low density, known as electron-hole liquid [11], which occurs when the interactions between the excitons are attractive; short lifetime of excitons in many systems, which prevents them from ever reaching a thermodynamic equilibrium; and density-dependent recombination[12, 13], which limits the maximum density in some systems.

Over the past decade there has been concerted effort to observe BEC of excitons in a semiconductor system engineered to overcome these problems. The system of indirect excitons in coupled quantum wells [14, 15] is in many ways an optimal system for observing BEC of excitons, and numerous experimental efforts [16, 17, 18, 19] have been directed toward this goal. In this system, carriers are trapped in a set of two identical, parallel quantum wells. When voltage is applied normal to the wells, the electrons are all drawn to one of the wells, while holes are all drawn to the other well, and “indirect” excitons form as bound pairs of spatially separated electrons and holes. This system has several properties which favor BEC. First, the lifetime of the excitons is greatly extended, in the present experiments up to 10 µs, compared to exciton-phonon scattering times in the range of tens of picoseconds up to nanoseconds, which allows them to reach thermodynamic equilibrium. Second, since the indirect excitons are aligned dipoles, they have an overall repulsive interaction, and electron-hole liquid cannot form [15]. Last, the quantum confinement in one direction means that the exciton gas is two-dimensional, and therefore the total number of excitons needed for BEC at a given temperature is much lower than in three dimensions. In typical experiments with bulk crystals, the upper limits of available laser intensities must be used, while in the quantum well experiments, the needed densities are easily reached with standard laser systems.

The greatest problem with coupled quantum well structures has been the presence of disorder [20], which arises during the growth process through a number of mechanisms, such as well width variation, disorder in the alloys used for some of the epitaxial layers, and impurities. Typical GaAs quantum well structures in the early 1990’s had inhomogeneous luminescence line broadening, which is a good measure of the disorder energy, of around
5 meV. If the excitons have kinetic energy less than the disorder energy, the excitons will mostly be localized in random energy minima; in other words, for disorder energy of 5 meV, at temperatures less than around 50 K most of the excitons will be unable to move. If the excitons are localized, one cannot treat them as a gas to which one can apply equilibrium thermodynamics of phase transitions. If the temperature is raised, the excitons can become ionized. Since the binding energy of excitons in GaAs quantum wells ranges from 5-10 meV, depending on the exact structure[21], a substantial fraction of the excitons will be ionized if $k_B T$ is large compared to a disorder energy of 5 meV. Also, the density which is needed for BEC increases with increasing $T$, so that raising the temperature of the excitons may mean that the density needed for BEC reaches the range at which the excitons dissociate due to collisions [22].

The quality of GaAs quantum well structures, however, has been steadily improving. One approach which substantially decreases the effect of disorder, and therefore increases the mobility, is to use wide quantum wells. Many previous studies have used narrow quantum wells of 50-60 Å GaAs, because the binding energy of the excitons is increased for narrower wells. The mobility in quantum wells is approximately proportional to the well width to the sixth power [23], however, which strongly favors wider wells. Another factor which favors wider quantum wells is the effect of well width on the interactions between the excitons. As shown by Lozovik and Berman [15], the interactions between excitons become completely repulsive for total electron-hole separation of three times the two-dimensional excitonic Bohr radius, which in GaAs is approximately 65 Å. For these reasons, for this study we have used a structure with two 100 ÅGaAs quantum wells, separated by a thin, 40 Å Al$_3$Ga$_7$As barrier, which has exciton binding energy at high electric field of 3.5 meV [24].

A second hurdle to overcome is the effect of a current of free charge carriers passing through the quantum wells. Because we apply voltage normal to the wells, there is always a current due to drift of carriers from the surrounding, doped substrate, which tunnel through the outer barriers confining the electrons and holes in the quantum wells. These carriers will tend to screen out the electron-hole Coulomb interaction, thus reducing the binding energy, and they will decrease the mobility of the excitons by random scattering with free carriers. In previous work [25, 26], the current of free carriers played a major role in the explanation [27, 28] of the mysterious effect of luminescence rings. To reduce the current
through our structure, we take employ three strategies. First, we use the highest possible barriers, Al$_{1.45}$Ga$_{0.55}$As. Second, we introduce into the barriers a superlattice of 60 Å GaAs wells, which act to trap free carriers moving across the barriers, following the approach of Dremin et al.[17]. Last, the excitons are created using a laser with wavelength nearly resonant with the single-well exciton energy. All of these approaches lead to a strong reduction in the current; for these samples the typical dark current is less than 1 µA/cm$^2$.

All of the above design features lead to a structure in which the indirect excitons have low disorder and high mobility. Figure 1 shows a time-integrated image of the indirect exciton luminescence, following a short (200 fs) laser pulse resonant with the single-well exciton energy, for the coupled quantum well system described above, immersed in liquid helium at $T = 2$ K. This image was taken by projecting an image of the sample onto the entrance slit of an imaging spectrometer with a CCD camera on the back focal plane, so that the horizontal axis corresponds to the position of the sample while the vertical axis corresponds to the luminescence photon energy. Although the laser is focused to a tight spot of 30 µm, the exciton luminescence is spread over hundreds of microns, due to the motion of the excitons. The bright, spectrally broad but spatially narrow line is luminescence from the doped GaAs substrate and capping layer, which are necessarily also excited by the laser. The substrate luminescence has short lifetime and therefore does not spread out spatially. The exact energy of the indirect excitons depends on the applied electric field, according to the quantum confined Stark effect [29]. In these experiments, the indirect exciton line is shifted by high electric field to an energy below the bulk GaAs band gap.

As seen in Figure 1, the indirect exciton energy is shifted upward in the center, where the density is highest, and is lower for locations away from the center, falling back to the unrenormalized value as the density of the excitons drops. This is consistent with the expected mean-field blue shift of bosons with repulsive interaction potential. As shown in Figure 2, the size of the exciton cloud becomes larger at higher densities, approximately proportional to the square root of the laser power, which also indicates that there is a pressure in the exciton gas due to the repulsive interactions between the excitons.

Figure 3 shows time-resolved luminescence for various distances from the excitation spot. The rise time of the luminescence is longer for spots further away from the excitation spot, consistent with the picture that the excitons move out from the excitation spot without
becoming localized. The lifetime of the excitons is 6.5 $\mu$s, in agreement with the calculated enhancement of the oscillator strength of the indirect excitons. In all cases the period between laser pulses was 4 $\mu$s, which means that some of the excitons created by each laser pulse will remain at the time of the succeeding pulse. Unlike previous experiments [25, 26], there are no intermediate dark states; the exciton cloud moves continuously from the laser spot outward.

We conclude that the excitons are delocalized as a free gas, traveling distances of hundreds of microns. This is an essential point, because the free motion of the excitons over long distances must be established if we are to believe that we observe a thermodynamic phase transition and not localized excitons.

BEC is not possible in two dimensions unless there is a confining potential. Therefore, we create a harmonic potential minimum in the plane of the excitons using the stress method described elsewhere [30]. One advantage of this method is that we can easily switch between a two dimensional, translationally invariant system and a confined system by removing or applying the external stress. Previous experiments [18, 20] have attempted to observe a Kosterlitz-Thouless superfluid transition in a translationally invariant system, but the hydrodynamics of a freely expanding superfluid are poorly understood. Alternatively, some previous work [17] has centered around accumulation of excitons in local minima created by disorder in the wells, but these minima are very small and their exact spatial profile is poorly mapped.

In order for the excitons to act as a gas confined in a potential minimum, the diffusion length of the excitons must be comparable to the equilibrium size of the exciton cloud in the trap. This is the case in our experiments for these high-mobility excitons. Figure 4(a) shows the exciton luminescence energy vs. position in the two-dimensional plane for a harmonic potential created by stress. The excitons were created on one side of the potential minimum, and they then flowed from the excitation spot in all directions toward the energy minimum. Although the potential energy is not strictly a harmonic potential, we can approximate the potential energy at the minimum as a harmonic potential $U(x) = \frac{1}{2}kx^2$, with an effective spring constant of $k = 31 \pm 3$ eV/cm$^2$. As seen in this figure, the excitons move a long distance from the creation spot, flowing distances comparable to the size of the harmonic potential trap. Again, this shows that the excitons are delocalized and flow freely in response
to the drift force created by the stress gradient. If we move the laser spot to the center of
the trap, the exciton cloud contracts into a small cloud at the bottom of the trap. Figure
4(b) shows an image made in the same way, but with the laser moved to the center of the
trap. Figure 4(c) shows the profile of the exciton luminescence intensity in the trap for
various times after the laser pulse. At early times the profile is a sum of the distribution
left over from the previous laser pulse and the newly deposited excitons with spatial width
approximately 30 µm. At late times, the exciton gas maintains a nearly constant size, which
shows that the trap acts to confine the exciton gas.

The interactions of the particles act to flatten out the external potential, due to the mean-
field, density-dependent blue shift discussed above. We can deduce this effective potential
if we assume that at late times, the spatial profile of the gas is given by $I(x) \propto e^{-U(x)/k_B T}$,
where $U(x)$ is the sum of the external potential and the internal potential due to interac-
tions. Figure 5 shows a plot of $U(x)/k_B T = -\ln I(x)$. In this case, the effective spring
constant, found by fitting the potential minimum to a harmonic potential, is approximately
1.7 eV/cm².

In principle, BEC in a harmonic potential minimum implies a spatial condensation in
the lowest eigenstate of the potential. The interactions between the excitons, however,
will cause the condensate to broaden spatially [31], and for strong interactions the size can
become comparable to the classical equilibrium distribution. As shown by Keeling et al.[32],
however, the in-plane momentum distribution of the condensate will differ strongly from
that of particles in excited states. This implies a strongly peaked angular distribution of
the emitted light. Essentially, if the condensate is in the lowest momentum state, then the
in-plane momentum is zero, and since the light emission process conserves the in-plane $k$-
vector, the light emission should correspond to a diffraction-limited beam emitted normal to
the plane in which the excitons are confined.

Figure 6 shows the angular distribution of the luminescence light emission from the quan-
tum well structure for several different powers. Typically, one thinks of keeping the density
constant and dropping the temperature to approach the BEC phase transition, but as men-
tioned above, the temperature of the excitons is not a control parameter, instead being
determined by the rate of phonon emission. Instead, one can approach the BEC phase tran-
sition by keeping all conditions the same and increasing the particle density, e.g. in our
case by simply turning up the laser power. The effective temperature of the excitons should remain constant, assuming the rate of phonon emission is constant. As seen in Figure 5, at low excitation power, the angular distribution is broad, corresponding to the maximum angular acceptance of our light collection system, while at high exciton density, a central peak appears. In other words, the light emission is in the form of a beam, which has angular width of 0.014 radians. This implies a source size of 70 µm, assuming diffraction-limited emission, i.e. \( \Delta \theta = 1.22 \lambda/d \). This is smaller than the equilibrium distribution size of 150 µm, but a reasonable estimate of the size of the high-density cloud initially created by the laser pulse. Preliminary time-resolved measurements indicate that the beam-like emission occurs primarily only at very short time delays after the laser pulse, consistent with the prediction [33] that the emission from the excitonic condensate should be superradiant, with very short lifetime.

This behavior is strikingly different from that of a nondegenerate exciton gas. Luminescence emission from excitons in quantum wells at \( T = 2 \) K is emitted in all directions. At this point, however, we cannot conclude that the angular peak definitely comes from Bose-Einstein condensation. Several facts appear inconsistent with this interpretation. First, the angular width of the peak does not vary strongly with density, although the width is larger than our experimental angular resolution of 0.5 degrees. Theory [32] predicts that the width of the peak should vary with density. Also, the spectrum of the light emitted from the condensate peak should be narrow compared to the spectrum from the rest of the gas, while we see just one, broad spectral peak.

To see whether BEC should be expected, we can compare the estimated density of particles to the theoretical prediction for condensation. The critical number of particles need for condensation in a harmonic potential in two dimensions, in the Stringari-Pitaveskii limit [34], is equal to

\[
N_c = \frac{1.6g(k_BT)^2}{(\hbar \omega_0)^2},
\]

where \( g \) is the spin degeneracy (equal to 4 for GaAs excitons), \( k_B \) is Boltzmann’s constant, and \( \omega_0 = \sqrt{k/m} \) is the natural frequency of the harmonic potential. For the effective spring constant of 31 eV/cm\(^2\), and the effective mass of the excitons in the plane of the quantum wells of 0.14\( m_0 \), where \( m_0 \) is the vacuum electron mass, this implies a critical number of excitons at \( T = 2 \)K of \( N = 10^6 \). The spring constant of the trap, however, is renormalized
by the interactions, as seen in Figure 5. For the effective spring constant of 1.7 eV/cm², the critical number is $2 \times 10^7$. If the temperature of the excitons does not cool all the way to 2 K, this number will be higher.

The critical threshold of 10 mW average power, seen in Figure 5, corresponds to $5 \times 10^{10}$ photons per pulse delivered to the sample, accounting for the efficiency of our optical system and 30% of the light reflected from the surface of the GaAs. The absorption length in GaAs for the laser wavelength of 797 nm is approximately 100 µm, while the total quantum well thickness is 240 Å = .024µm, which means that the fraction of photons absorbed in the quantum well structure is approximately $(.024)/(100) = 2.4 \times 10^{-4}$. This implies approximately $1.2 \times 10^7$ absorbed photons. This is comparable to, but below, the critical number for BEC in the trap estimated above for $T = 2$ K.

One explanation for the angular peak is that lasing actually occurs during the intense pump pulse which creates the excitons. This is not expected, since the rate of photon emission is so slow. Alternatively, one can take note of the fact that immediately after the laser pulse, the volume of the newly created excitons is much less than the quasiequilibrium size, and therefore the above equilibrium estimate may not apply. While the exciton gas at early time has relatively small volume, it may exceed the critical density for condensation.

In conclusion, we have at our disposal a new system with highly mobile excitons with long lifetime, which we can trap in a harmonic potential, and we can observe the emitted light with spatial, spectral, temporal, and angular resolution. Our estimates indicate that the densities of excitons are either at or just below the expected critical density for condensation. A beamlike emission is observed, which is either due to lasing or possibly nonequilibrium condensation of the excitons.

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FIG. 1: Time-integrated image of the luminescence from the indirect excitons at $T = 2K$, taken using an imaging spectrometer so that the vertical axis corresponds to the energy of the emitted photons, while the horizontal axis corresponds to the position in the plane of the quantum wells. The central, spatially narrow but spectrally broad line corresponds to light emission from the doped GaAs substrate and capping layers, while the spatially broad but spectrally narrow line corresponds to the emission from the indirect excitons, of interest in this study.
FIG. 2: Full-width at half-maximum of the exciton cloud as a function of average laser excitation power.
FIG. 3: Luminescence intensity as a function of time, for various distances $x$ from the excitation spot, under the same conditions as Figure 1. The position in the plane is offset by the same value of $y = 50\mu m$ for all the curves, to reduce the contribution of the light emission from the substrate and capping layer.
FIG. 4: a) Exciton luminescence energy as a function of position, when the laser excitation spot is located near to a potential minimum created by externally applied stress. The image is recorded in the same way as Figure 1. b) The same conditions, but the laser spot moved to the center of the potential minimum. The laser excitation spot has been translated 50 μm in the perpendicular direction in the plane of the excitons, to reduce the light from the substrate and capping layer. c) The intensity of the luminescence as a function of position, for various time delays after the laser pulse, under the same conditions as (b). Black curve: $t = 20$ ns. Blue curve: $t = 1.6$ μs. Red curve: $t = 3.6$ μs.
FIG. 5: The effective potential felt by the excitons, deduced from the intensity distribution in quasiequilibrium at $t = 3.6 \mu s$, shown in Fig. 4(c).
FIG. 6: Angular distribution of the light emission from the indirect excitons, for various laser powers. The angular width of the low-power emission corresponds to the response of our light collection system to a uniform source; i.e., our system has 9 degrees total angular acceptance. The angular resolution of our system is 0.5 degrees.