

# Long-range transport in excitonic dark states in coupled quantum wells

D. Snoke<sup>1</sup>, S. Denev<sup>1</sup>, Y. Liu<sup>1</sup>, L. Pfeiffer<sup>2</sup> and K. West<sup>2</sup>

<sup>1</sup>*Department of Physics and Astronomy, University of Pittsburgh, 3841 O'Hara St., Pittsburgh, PA 15260*

<sup>2</sup>*Bell Labs, Lucent Technologies, 700 Mountain Ave., Murray Hill, NJ 07974-0636*

In the past ten years, the system of coupled quantum wells has emerged as a promising candidate for experiments on Bose condensation of excitons, with numerous theoretical [1–6] and experimental [7–12] studies aimed at demonstration of this effect. One of the issues driving these studies is the possibility of long-range coherent transport of excitons. Excitons in quantum wells typically diffuse only a few microns from the spot where they are generated by a laser pulse; their diffusion is limited by their lifetime (typically a few nanoseconds) and by scattering due to disorder in the well structure. We report here a new effect by which luminescence from excitons appears hundreds of microns away from the laser spot. This luminescence appears in a ring around the laser spot; almost no luminescence appears in the region between the laser spot and the ring. We examine whether this effect may be related to macroscopic coherence due to Bose condensation of excitons.

Coupled quantum wells are attractive for Bose condensation of excitons because the lifetime of the excitons can be extended by application of an electric field. A typical band structure is shown in Fig. 1. When electric field is applied normal to the planes of the wells, electrons are confined to one well and holes are confined in the adjacent well. Because the electron and hole wavefunctions in the two wells have little overlap, excitons in this type of

structure have much longer lifetime, up to 100 ns [13]. Excitons formed from an electron in one well and a hole in the adjacent well are known as “indirect” excitons. Because the electric field shifts the electron and hole states to lower energy, in what is known as the “quantum confined Stark effect” [14,15], it is easy to identify the indirect exciton luminescence, since only the indirect exciton luminescence shifts to lower energy with increasing electric field.

Our present studies have focused on InGaAs coupled wells which have a confined state below the GaAs substrate on which they are grown. When this system is illuminated with red light (630 nm) well above the band gap of the GaAs substrate, carriers generated in the substrate will fall into excitonic states in the coupled quantum wells, which are the lowest excited state in the system. This is therefore a very efficient way of creating a high density of excitons in the quantum wells. At very high excitation density, the carriers screen out the electric field, leading to a blue shift of the exciton luminescence as seen in Ref. [16], but there exists a regime of density in which the indirect exciton luminescence does not shift substantially with excitation density.

In this regime we find that above a critical excitation density, the indirect exciton luminescence exhibits a ring structure centered on the laser spot. Fig. 2 shows typical images from our experimental data. An image of the sample is projected onto the entrance slot of an imaging spectrometer, which preserves the spatial information along the slit while spectrally dispersing the light in the perpendicular direction. These images from an intensified CCD camera on the back of the spectrometer therefore give the spectrum of the exciton luminescence as a function of position. The ring appears as two spots on these images. As seen in Fig. 2, the radius of the ring depends on the laser excitation density. The radius of the ring can also be changed by keeping the laser intensity constant and adjusting the applied voltage across the wells.

As seen in these images, the ring appears *many hundreds of microns distant from the laser spot*, while the intervening region is dark. We have confirmed that the structure is a ring centered on the laser spot by moving the laser spot; when the laser spot is moved upward, the ring follows the laser, remaining centered on it, and when the laser spot is

moved to the side, only the two spots from the ring structure are seen in the CCD images, with decreasing separation until they merge when the edge of the ring is projected onto the entrance slit of the spectrometer.

The most surprising aspect of these images is that no luminescence appears in the space between the laser spot and the ring for hundreds of microns. This implies that the excitons must travel in some dark state until they reach some critical distance, at which they collectively revert to luminescing states. This type of behavior is unprecedented in other excitonic systems.

One one hand, it would seem natural to interpret this effect as arising from Bose coherent effects of the excitons, but since this effect would be a retrodiction, not a prediction, of any present theory of Bose coherence of excitons, we do well to ask what other tests show. One obvious test is to determine the temperature of the critical density. The critical temperature for Bose superfluidity depends on the density; alternatively, one can speak of the critical density at a given temperature. We have observed the ring structure at a series of different temperatures and determined the critical excitation density at which the ring appears for each temperature. The excitation density is simply the number of photons per laser pulse divided by the area of the laser spot. Since the laser at this wavelength is essentially 100% absorbed by the substrate near the quantum wells, the excitation density will be approximately the same as the exciton density in the wells if the exciton lifetime is less than the period between laser pulses; if the exciton lifetime is much longer than this period, the exciton density can greatly exceed this value.

The results for the critical excitation density as a function of temperature are shown in Fig. 3. The critical density is defined by the point at which the ring radius is zero, as shown in the inset of Fig. 3. In our coupled quantum well structure, the binding energy of the indirect excitons is approximately 10 meV. At  $T = 90$  K, free electrons and holes coexist with the excitons, but we still see the ring structure at high laser excitation density. No free electrons and holes are seen in the dark region between the central spot and the ring, which indicates that the transport occurs entirely in dark states even at these temperatures.

For a Kosterlitz-Thouless transition to a superfluid state in two dimensions, the critical density is expected to be directly proportional to the temperature [17], equal to

$$\sigma_c = \frac{2k_B T m}{\pi \hbar^2}, \quad (1)$$

which gives a critical density of  $8 \times 10^{10} \text{ cm}^{-2}$  for an effective exciton mass of  $m = 0.12m_0$  (the expected in-plane effective mass of excitons in InGaAs quantum wells) and  $T = 10 \text{ K}$ . As seen in Fig. 3, the observed critical excitation densities are approximately proportional to the square of the temperature, instead of linear. This would tend to argue against the possibility of a Bose coherent transition. On the other hand, for Bose condensation in a potential minimum, the critical number for condensation is proportional to the square of the temperature [17], and the critical number of excitons can be far less, depending on the properties of the potential minimum. Butov et al. [10] have argued that indirect excitons in this type of coupled quantum well can be trapped in a local minimum created by random fluctuations in the quantum well composition. Alternatively, one could argue that many-body effects of the exciton gas create a potential minimum wherever the excitons are, e.g. a local reduction of the band gap due to heating of the lattice. There is evidence for this in the fact that the indirect exciton luminescence at the laser spot always appears slightly lower in energy than the luminescence from the ring. As seen in Fig. 2, the indirect exciton luminescence at the laser spot is around 1 meV lower in energy. This fact raises the question, however, of why the Bose-condensed excitons would leave the trap to form the ring at higher energy. The standard prediction for Bose condensation in a trap is that the condensate will form in the center, at the point of lowest energy.

One possible mechanism for the outward expansion is that a “phonon wind” pushes the excitons out of the central region. It has long been known [18–20] that a laser with photon energy well above the band gap can generate a nonequilibrium distribution of phonons that act like a wind to push excitons. Normally, however, such a wind does not create a well-defined ring structure in a time-integrated image; the excitons emit luminescence continuously as they move. A phonon wind should also be much less efficient at high

temperature.

A second test is to ask whether time resolving the luminescence gives results consistent with migration of the excitons from the laser spot to the ring. At typical exciton velocities at low temperature on the order of  $\mu\text{m}/\text{ns}$ , excitons should take hundreds of nanosecond to travel hundreds of microns. We have performed time-resolved studies of the rings, using time-resolved single photon counting with 40-ps resolution and a cavity-dumped, mode-locked pulsed laser with 2-ps pulse duration. As seen in Fig. 4(a), the indirect exciton luminescence at the laser spot has fairly short lifetime, around 3 ns, while the luminescence from the ring has long lifetime. By changing the repetition rate of the laser using a cavity dumper to give very long period between the pulses, we can directly measure the long-time behavior of the excitons at the ring. Fig. 4(b) shows the intensity of the ring as a function of time after a laser pulse, for three different distances of the ring from the central spot. As seen in this figure, the rise time of the luminescence is longer when the ring is further from the central spot, consistent with the greater distance traveled in dark states, and the exciton lifetime is also consistent with travel over such long distances. The rise times shown in Fig. 4(b) correspond to a velocity of the excitons of approximately  $1.5 \times 10^6$  cm/s, which is reasonable for drift-like motion of excitons but three times faster than the speed of sound in GaAs, which would seem to rule out a phonon wind mechanism. These data also rule out the possibility of a radiative reabsorption effect, by which one could imagine that the ring occurs not by actual migration of the excitons, but by recombination into photons which are then reabsorbed at further away in the well. Radiative reabsorption would give a rise time and lifetime the same as that of the luminescence in the central spot.

In general, there are two possibilities for dark states in GaAs quantum wells. Hot excitons will not emit luminescence if they have energy greater the line width of the direct recombination luminescence line, which is less than 1 meV. Since excitons cool to the lattice temperature within a few nanoseconds, however, it is unlikely that they all remain in high-energy states, with no emission from the low-energy states, for hundreds of nanoseconds. The excitons also have dark states which are nearly degenerate with the bright states [21]

but may lie slightly lower in energy. A condensate in one of these states could prevent luminescence until the excitons revert to the normal state.

It is worth asking what is different in these experiments from previous experiments, and why this ring has not been seen before in similar structures. There are two primary differences in these new experiments. First, the quality of these new quantum well structures is very high. The full width at half maximum of the luminescence line at low temperature and low excitation density in these InGaAs quantum well samples ranges from 0.7 to 1.2 meV, which is very low compared to typical narrow quantum well structures. Second, many previous experiments have used near-resonant excitation, under the assumption that the exciton temperature would be lower with less excess photon energy. In these experiments, we use red (ca. 630 nm) light which is well above the band gap of the substrate. The absorption of the laser light in this case is nearly 100%, compared to around 5% for near-resonant excitation. Most of the light is absorbed in the substrate, after which free carriers can fall into the quantum wells. It may be that producing a large number of free carriers in this way is an ideal method for achieving a high density of excitons; alternatively, the high-energy carriers may lead to novel screening effects that have nothing to do with Bose coherence. As discussed above, the red laser light may also have the effect of creating a phonon wind and self trapping due to heating of the lattice. Finally, it is possible that this effect has occurred but not been observed before, because the luminescence from the ring structure is weak and would not be observed without an intensified imaging system.

Because of the unsettled issues discussed above, it is not clear whether this effect is related to Bose coherence. In any case, however, the transport of excitonic signal over such large distances, with a dark intermediate state, is a novel effect which deserves further study. With exciton motion over such long distances, it is possible to imagine “excitonic circuits,” in which signals are carried by excitonic degrees of freedom, similar to “spintronics,” in which signal is carried by spin.

## REFERENCES

- [1] Zhu, X., Littlewood, P.B., Hybertson, M.S., & Rice, T.M. Exciton condensate in semiconductor quantum well structures. *Phys. Rev. Lett.* **74**, 1633-1636 (1995).
- [2] Fernández-Rossier, J. & Tejedor, C. Spin degree of freedom in two-dimensional condensates. *Phys. Rev. Lett.* **78**, 4809-4812 (1997).
- [3] Lozovik, Yu.E., & Birman, O.L. Phase transitions in a system of spatially separated electrons and holes. *JETP* **84**, 1027-1035 (1997).
- [4] Yudson, V.I. Charged “few-electron– single spatially separated hole” complexes in a double quantum well near a metal plate. *Phys. Rev. B* **77**, 1564-1567 (1996).
- [5] Iida, T. & Tsubota, M. Order formation and superfluidity of excitons in type-II semiconductor quantum wells. *Phys. Rev. B* **60**, 5802-5810 (1999).
- [6] Dzubenko, A.B., & Yablonskii, A.L. Intrawell and interwell magnetoexcitons in  $\text{In}_x\text{Ga}_{1-x}$  coupled double quantum wells. *Phys. Rev. B* **53**, 16355-16364 (1996).
- [7] Larionov, A.V., & Timofeev, V.B. Condensation of interwell excitons in GaAs/AlGaAs double quantum wells. *JETP Lett.* **73**, 342-350 (2001).
- [8] Krivolapchuk, V.V., Moskalenko, E.S., Zhmodikov, A.L., Cheng, T.S. & Foxon, C.T. Collective properties of spatially indirect excitons in asymmetric GaAs/AlGaAs double quantum wells. *Solid State Comm.* **111**, 49-54 (1999).
- [9] Butov, L.V. & Finin, A.I. Anomalous transport and luminescence of indirect excitons in AlAs/GaAs coupled quantum wells as evidence for exciton condensation. *Phys. Rev. B* **58**, 1980-2000 (1998).
- [10] Butov, L.V., Lai, C.W., Ivanov, A.L., Gossard, A. C., & Chemla, D. S. Towards Bose-Einstein condensation of excitons in potential traps. *Nature* **417** 47-52 (May 02, 2002).
- [11] Negoita, V., Snoke, D.W., & Eberl, K.. Stretching quantum wells: a method for trapping

- free carriers in GaAs heterostructures,” *Applied Phys. Lett.* **75**, 2059-2061 (1999).
- [12] Negoita, V., Hackworth, D., Snoke, D.W., & Eberl, K. Sub-Hz spectral fluctuations from high-density excitons in biased coupled quantum wells. *Optics Lett.* **25**, 572-574 (2000).
- [13] Negoita, V., Snoke, D.W., & Eberl, K. Harmonic potential traps for indirect excitons in coupled quantum wells. *Phys. Rev. B* **60**, 2661-2669 (1999).
- [14] Fox, A.M., Miller, D.A.B., Livescu, G., Cunningham, J.E., & Yan, W.Y. Excitonic effects in coupled quantum wells. *Phys. Rev. B* **44**, 6231-6242 (1991).
- [15] Kato, Y., Takahashi, Y., Fukatsu, S., Shiraki, Y., & Ito, R. Observation of the Stark effect in coupled quantum wells by electroluminescence and circularly polarized photoluminescence excitation spectroscopy. *J. Appl. Phys.* **75**, 7476-7481 (1994).
- [16] Negoita, V., Snoke, D.W., & Eberl, K. Huge density-dependent blueshift of indirect excitons in biased coupled quantum wells. *Phys. Rev. B* **61**, 2779-2783 (2000).
- [17] Moskalenko, S.A. and Snoke, D.W. *Bose-Einstein Condensation of Excitons and Biexcitons*. (Cambridge University Press, 2000).
- [18] Bagaev, V.S., Keldysh, L.V., Sibeldin, N.N., & Tsvetkov, V.A. Phonon wind drag of excitons and electron-hole drops. *Soviet-Physics JETP* **70**, 702-716 (1976).
- [19] Hensel, J.C., & Dynes, R.C. Interaction of electron-hole drops with ballistic phonons in heat pulses: The phonon wind. *Phys. Rev. Lett.* **39**, 969-972 (1977).
- [20] Greenstein, M. and Wolfe, J.P. Anisotropy in the shape of the electron-hole-droplet cloud in germanium. *Phys. Rev. Lett.* **41**, 715-719 (1978).
- [21] Snoke, D.W., Rühle, W.W., Köhler, K., & Ploog, K. Spin flip of excitons in GaAs quantum wells. *Phys. Rev. B* **55**, 13789-13794 (1997).

**Acknowledgements.** This work has been supported by the National Science Foundation and by the Department of Energy. We thank Viorel Negoita for early contributions to these experiments.

FIG 1. The band structure of the coupled quantum wells in the presence of electric field. Indirect excitons are made from an electron in one well and a hole in the adjacent well. (The dashed line indicates the optical transition which gives rise to the luminescence from indirect excitons.)

FIG 2. Spectral images of the luminescence from indirect excitons for various laser powers. a) 1.7 mW, b) 2.0 mW, c) 2.5 mW. The ring around the laser spot is seen in these figures as a pair of dots equidistant from the intense, central spot, which appears at the focal spot of the laser beam which creates the excitons. The horizontal axis gives the wavelength of the luminescence. As seen in this figure, the ring appears at slightly shorter wavelength (higher energy) than the luminescence from the excitons at the laser focus.

FIG. 3. The critical excitation density for appearance of the luminescence ring, as a function of the bath temperature in a variable-temperature cryostat. The solid line is a fit to a  $T^2$  dependence. Inset: The ring radius vs. excitation density. The critical threshold is defined as the point at which the ring merges with the the central spot, which occurs in this case at  $x = 160\mu\text{m}$ . The solid line is a linear fit of the data. At higher excitation density, the radius saturates and even decreases with increasing power.

FIG. 4. a) Time-resolved streak image of the indirect exciton luminescence profile. The center of the laser spot, corresponding to  $x = 0$  has short lifetime, while the ring (at  $x = 400\mu\text{m}$ ) has long lifetime. The laser repetition period in this case is 13 ns. As seen in this figure, no light appears in the region between the central spot and the ring at any time, except for scattered laser light during the laser pulses. b) Intensity of the luminescence from the ring as a function of time delay after the laser pulse, for two different ring radii, for a laser repetition period of 260 ns. The luminescence in the case of the  $1311\text{-}\mu\text{m}$  ring is offset for clarity.

FIGURES

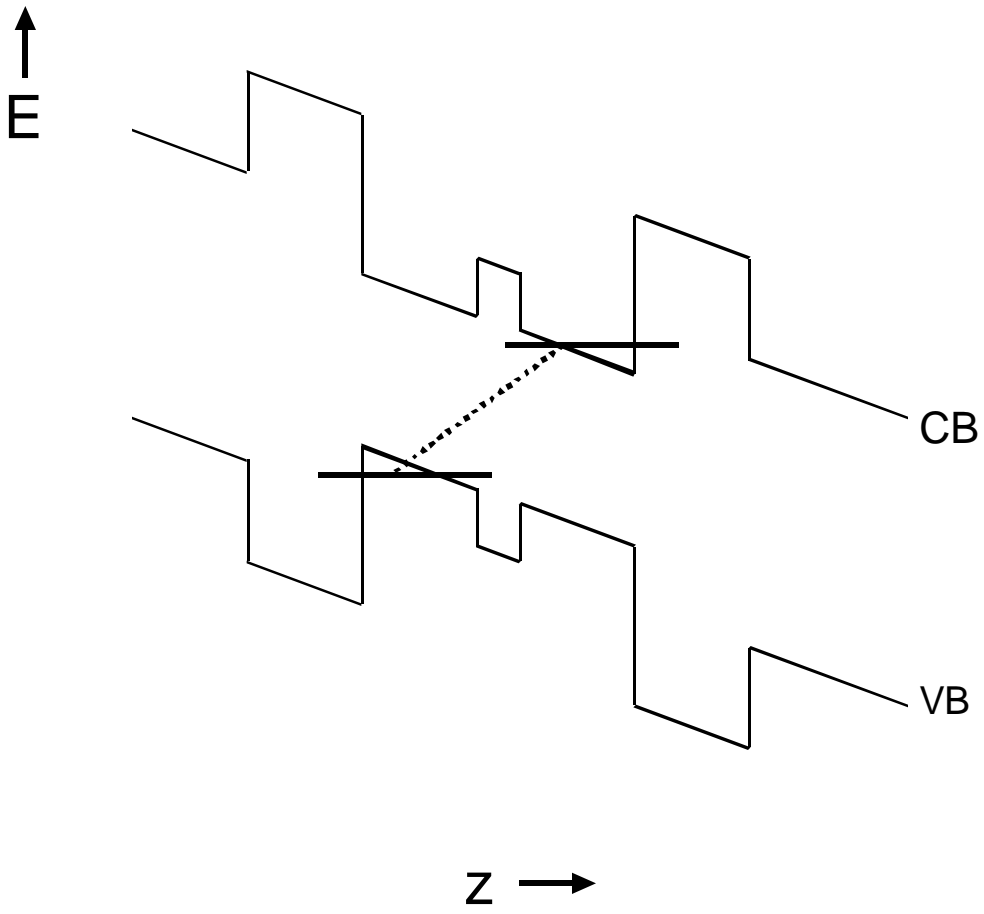


FIG. 1.

FIG. 2.

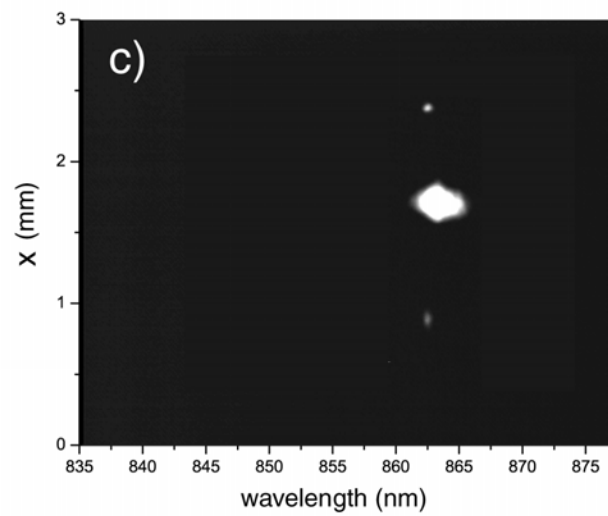
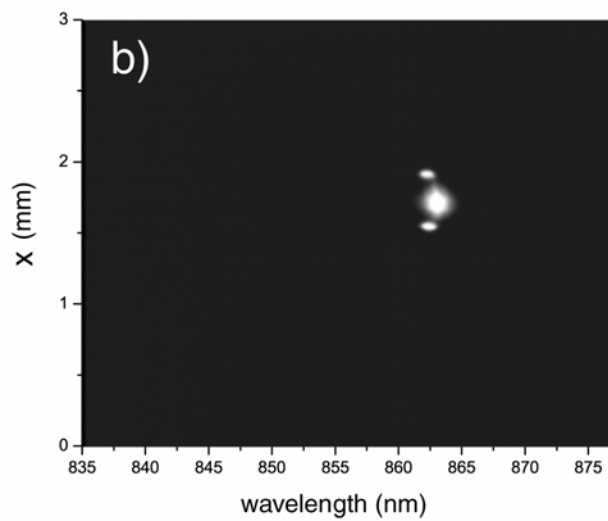
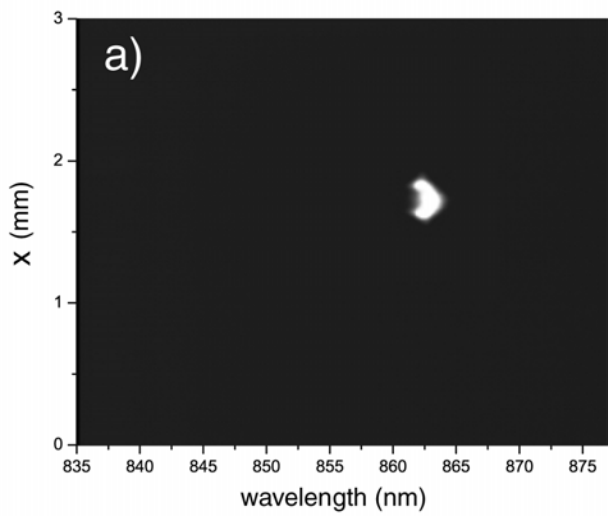


FIG. 3.

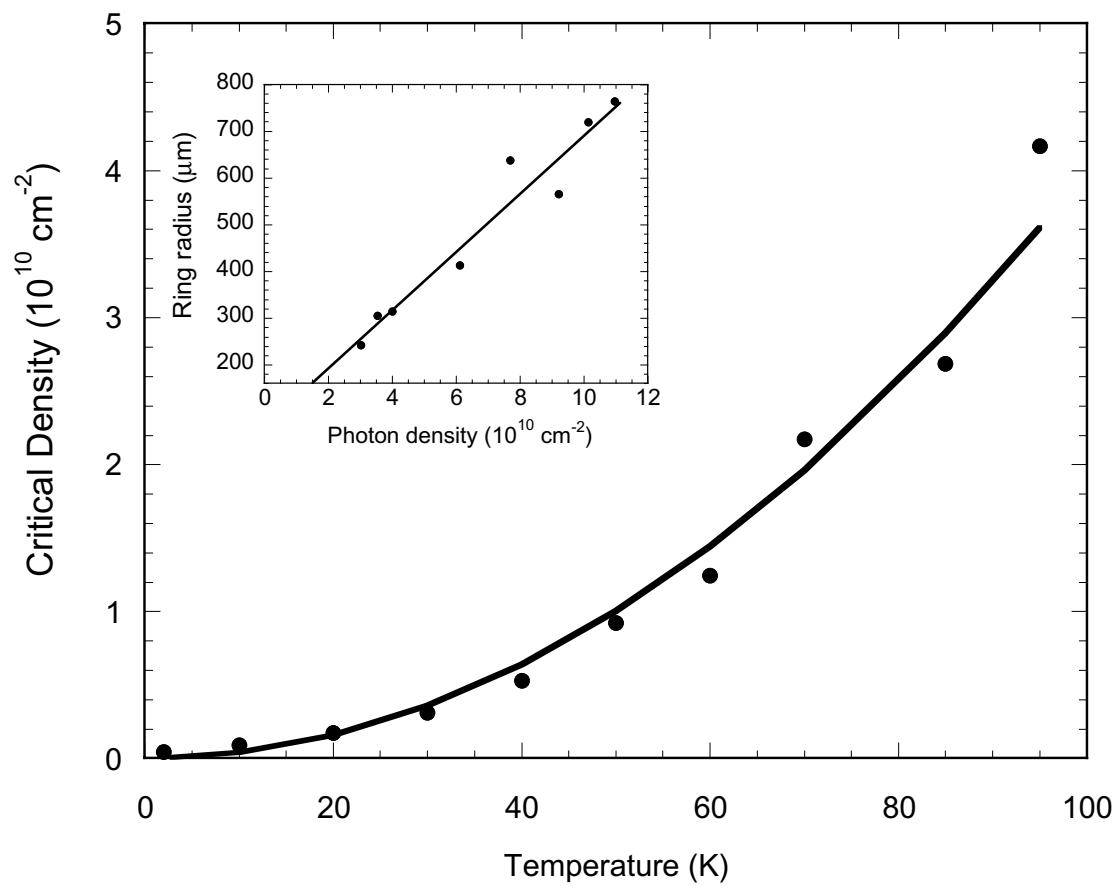


FIG. 4(a).

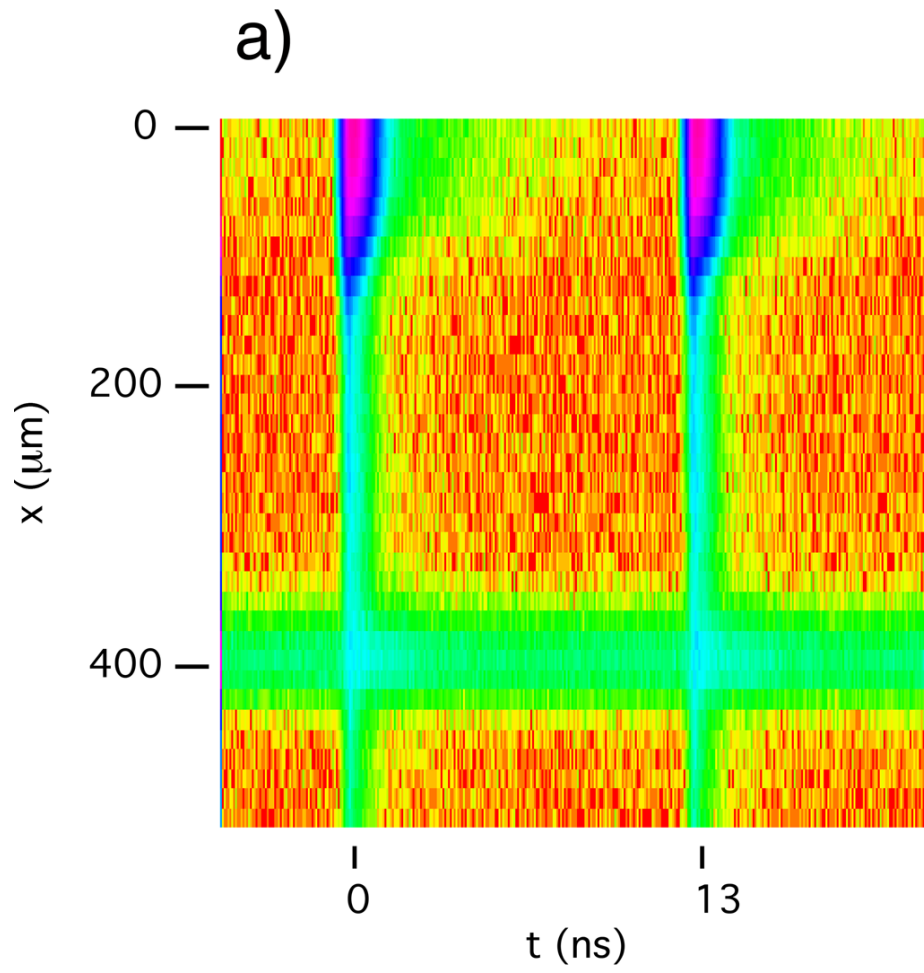


FIG. 4(b).

