Bosonic stimulation of cold 1s excitons into a harmonic potential minimum in Cu$_2$O

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Density distribution of cold exciton clouds generated into a strain-induced potential well by two-photon excitation in Cu$_2$O is studied at 2 K. We find that an anomalous spike, which can be interpreted as accumulation of the excitons into the ground state, emerges at the potential minimum. The accumulation can be due to stimulated scattering of cold excitons, mediated by acoustic phonon emission. Possibility of the formation of the thermodynamic Bose-Einstein condensate of paraexcitons has been discussed. PACS numbers: 71.35.Lk, 78.35.-y

Before the first realization of atomic Bose-Einstein condensation (BEC) in 1995 [1], excitons in semiconductors were thought to be one of the most favorable candidates for realizing BEC [2]. It has long been believed that excitons in Cu$_2$O is the best system to pursue such a phase transition in a three-dimensional system, because of their long lifetime derived from the dipole forbidden gap; the orthoexcitons couple with photons only via quadrupole interaction, and paraexcitons are basically inactive for optical transitions, thereby leading to a long lifetime.

However, the long lifetime of the excitons in Cu$_2$O allows them to expand in free crystals, and means that their recombination rate is low. The former reduces the exciton density against the BEC, and the latter makes it difficult to monitor exciton densities by optical means. In order to overcome these unfavorable situations, we employed inhomogeneous strain that creates potential wells and accelerates the direct recombination rate. We have been studying kinetics of a cold exciton system in a potential well, by using two-photon excitation. In this regime, orthoexcitons are created near the local momentum zero [3, 4]. In this communication, we report on a novel phenomenon showing stimulated scattering of excitons; this can be mediated by acoustic phonon emission.

A natural Cu$_2$O crystal cut into a 3 mm-cube was pushed against a glass lens of curvature $R = 7.78$ mm to create harmonic potential wells (see the inset of Fig. 1). Solid lines in Fig. 1 show calculated potential shapes for the four lowest exciton states in Cu$_2$O, i.e. upper-, middle- and lower-branch orthoexcitons and paraexcitons. It is evident that middle- and lower-branch orthoexcitons as well as the paraexcitons become trapped states while the upper-branch orthoexcitons form an untrapped state. The trapping potential shapes are approximately harmonic around the center of the wells. An infrared laser beam from a color-center laser (Solar, LF151) pumped by a Q-switched YAG laser (SantaFe Laser, C-140) was used to inject orthoexcitons into a potential well. The repetition rate and the duration of the laser light were 400 Hz and 12 ns, respectively. The two-photon excitation energy of the laser light was tuned slightly below the orthoexciton resonance under zero stress, so that middle- and lower-branch orthoexcitons were generated into the trap [5]. The luminescence was viewed along the stress axis through a prism mounted below the crystal. A $\times 20$ magnified image of the luminous cloud was detected by an ICCD camera system (LaVision, Picostar HR12) after being passed through a filter which cuts off the infrared scattering light. By inserting a spectrometer before the ICCD camera system, we confirmed that luminescence light consists of intrin-
FIG. 2: Time evolution of the spatial profile of the exciton cloud pumped at a rim of a potential well. The baseline for each profile has been shifted for clarity. A sharp spike emerges at a rim of a potential well. The baseline for the spatial profile at a rim of a potential well.

Part (a) and (b) show growing and decaying of the exciton density, with time interval of 2 ns. Three major peaks are seen in a range between $y = 200 \mu m$ and $y = 400 \mu m$. The intensity of the side peaks was almost proportional to the temporal profile of the laser pulse, and to the square of the power density of the laser light. On the other hand, the central peak shows rapid growth even after the incidence of the laser light. The gate width was set at 4 ns, and the power of the incident light was $I = 9$ mW on the sample surface.

In order to understand the origin of the side peaks and of the anomalous spike in between, the spatial profile at $t = 10$ ns is superimposed to the potential shapes in Fig. 1. The position of the center of the well for middle-branch orthoexcitons is almost the same as that for paraexcitons. Therefore, judging from its position, the origin of the spike can be middle-branch orthoexcitons, paraexcitons, or both. Unfortunately, however, we could not obtain spectrally resolved data in the present experiment, because of the weakness of the signal. Dashed arrows in Fig. 1 shows a possible mechanism for the emergence of the spike: the orthoexciton-paraexciton downconversion occurs in the injection spot at a rim (the arrow labeled 1), and then paraexcitons at the rim are scattered into the center of the well (the arrow labeled 2). The energy transfer necessary for this scattering process is inferred to be 0.6 meV, in agreement with the energy position where the acoustic (LA)-phonon dispersion crosses the exciton dispersion. Therefore, the scattering can be mediated by acoustic phonon emission. In fact, we found that the central spike appears only when the laser beam traverses the center of the well, and when the two-photon excitation energy is set at $\Delta E = -5.03 \pm 0.25$ meV. Considering the timescale of the downconversion rate, 3 ns [5], and the energy transfer, 1.0 meV or more, necessary for the scattering of orthoexcitons from the rim to their potential minimum, condensation of the middle-branch orthoexcitons is not likely. To conclude, the spike marked C is most likely to be due to paraexcitons accumulated in the ground state at the center of the potential well.

The role of the acoustic phonon in the accumulation of the condensate has been discussed in Refs. [2, 3]. The building time has been estimated as a few nanoseconds. The observed delay is in quantitative agreement with this prediction, provided that the central spike is due to a condensate. Using the measured size of the central spike, the phase-space density of paraexcitons was estimated as $\geq 1$. A more detailed measurement of the building time of the spike deserves future study. Confirmation with higher spectral resolution, which enables measurements of the size of the ground-state wavefunction, is in progress.

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[1] M.H. Anderson, J.R. Ensher, M.R. Matthews, C.E. Wiemann, and E.A. Cornell, Science 269 (1995) 198.

[2] Bose-Einstein Condensation, A. Griffin, D.W. Snoke, and S. Stringari, eds., (Cambridge University Press, Cam-
bridge, 1995).

[3] N. Naka and N. Nagasawa, *Phys. Rev. B* **65** (2002) 075209.

[4] N. Naka and N. Nagasawa, *Phys. Rev. B* **65** (2002) 245203.

[5] J.S. Weiner, N. Caswell, P.Y. Yu, and A. Mysyrowicz, *Solid State Commun.* **46** (1983) 105.

[6] O.M. Schmitt, D.B. Tran Thoai, L. Bányai, P. Gartner, and H. Haug, *Phys. Rev. Lett.* **86** (2001) 3839.

[7] A. Schmitt, L. Bányai, and H. Haug, *Phys. Rev. B* **63** (2001) 205113.

[8] The two-photon excitation of paraexcitons is basically possible, but is not practical to generate a high-density gas of excitons [4].