Coherent Quantum Control of Excitons at Ultracold and High Density in Cu_2O with Phase Manipulated Pulses

Takuro Ideguchi,[1] Kosuke Yoshioka,[1] Andre Mysyrowicz,[2] and Makoto Kuwata-Gonokami[1,*]

[1]*Department of Applied Physics, The University of Tokyo, and CREST-JST, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*

[2]*Laboratoire d'Optique Appliquee, ENSTA, Ecole Polytechnique, Palaiseau, France*

(Received 15 February 2008; published 10 June 2008)

By phase manipulation of a short laser pulse, it is possible to selectively generate ultracold excitons in a two-photon process while quenching the multiphoton excitation of hot electrons and holes. We show how this technique allows us to suppress the heating of $n = 1$ orthoexcitons in Cu_2O at high density. Using a laser pulse having an energy of a few μJ and duration of 100 fs, we are thus able to produce a cold exciton gas up to a density of 10^{15} cm^{-3}.

DOI: 10.1103/PhysRevLett.100.233001 PACS numbers: 32.80.Qk, 32.80.Wr, 42.65.Re, 71.35.Lk

Phase and amplitude optical pulse shaping based on Fourier plane manipulation has been developed during the last decade [1] and applied to the control of optical transition in atomic or molecular gases [2,3] and large organic molecular systems [4,5]. The use of tailored pulses in condensed media is a challenging problem because of the fast dephasing processes at hand and the complexity of a situation where many transitions can occur simultaneously. In this Letter, we report on the demonstration of the coherent quantum control of excitons by elementary electronic excitation of semiconductors, using tailored femtosecond pulses.

The selected system is Cu_2O, a material in which excitons are a fascinating subject of study. The $n = 1$ excitons of the yellow series have a large binding energy (~ 150 meV), making them particularly stable against thermal or collision dissociation. Their radiative recombination is forbidden in the dipole approximation, conferring them a long lifetime [6]. Because of the electron-hole exchange interaction, the $n = 1$ excitons split into an optically inactive, singly degenerate Γ_2^+ paraexciton and triply degenerate Γ_5^+ dipole forbidden orthoexciton. Yellow $n = 1$ excitons in Cu_2O are therefore prime candidates for the observation of quasiequilibrium Bose-Einstein condensation (BEC) [7,8].

There are several effects which make it difficult to reach BEC of orthoexcitons by photo-excitation. If one produces orthoexcitons by nonresonant optical pumping, in the band-to-band continuum or in the phonon-assisted excitonic absorption edge below the band gap, one readily obtains a high density of the gas with a high degree of quantum degeneracy. However, there is also a concomitant heating of the gas, so that the ratio $-\mu/k_BT$ approaches 0 only asymptotically, preventing a large fraction of orthoexcitons to condense in the ground state [9] (μ is the internal chemical potential of the excitons). The excess energy of the initially created electron pairs or excitons leads to an effective temperature of the excitons higher than the lattice temperature. Resonant two-photon excitation [10] avoids the transfer of excess energy at creation, and with short pulses one can directly generate a low density of excitons into a super cooled state below the lattice temperature and thus prepare excitonic particles into a state with high phase space occupation before thermalization [11]. At densities below 10^{12} cm^{-3}, this ensemble of super cooled excitons can keep the acquired two-photon coherence over 250 ps [12]. Unfortunately, at a higher density excitation, a sudden heating of the gas is observed as shown below, the origin of which has remained a subject of controversy.

The technique of coherent quantum control with tailored pulses allows to clarify the mechanism responsible for this heating of excitons at intense excitation. We demonstrate that it is possible to suppress it to a large extent by simple manipulation of the optical pulses.

To understand the heating mechanism we refer to Fig. 1, where only the relevant exciton states coming into play are shown. Ascending arrows show incoming photons which give their energy to the crystal during the excitation process. As discussed below, a three-photon band-to-band

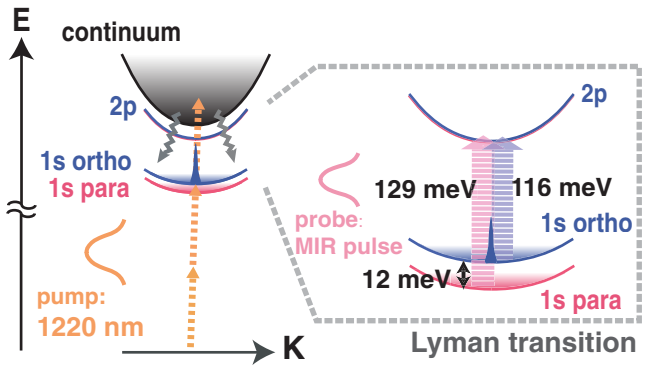

FIG. 1 (color online). Schematic description of the optical processes involved in the experiments. The infrared pulses tuned to two-photon resonance of 1s orthoexcitons directly excite 1s excitons near a zero kinetic energy and free electrons and holes via band-to-band continuum transition. The inset shows the probing scheme of exciton Lyman transitions.

transition which produces free electron-hole pairs can be identified as the lowest order process which transfers extra energy to the excitons. When coupling into excitons, these carriers release their binding energy in the form of kinetic energy that is shared between excitons during collisions, raising their temperature. This heating process becomes more and more important at higher laser intensities, I, since the rate of production of hot electron-hole pairs is proportional to I^3.

The following measurements support our interpretation. An infrared pulse tuned to the spectral region corresponding to the Lyman transition $n = 1$ to $n = 2$ measures the density of excitons and their temperature. The density of excitons present in the crystal is extracted from the magnitude of the integrated absorption line of this Lyman transition, the temperature from its line shape. The $1s$-$2p$ transition is described by the following equation:

$$\int_{1s\text{-}2p} \Delta\alpha(E)dE = \frac{n_{1s} 4\pi^2 E_{1s\text{-}2p} |\mu_{1s\text{-}2p}|^2}{\hbar c\sqrt{\varepsilon_b}}, \qquad (1)$$

where $E_{1s\text{-}2p}$, $\mu_{1s\text{-}2p}$ and ϵ_b are the $1s$-$2p$ transition energy, dipole moment and background dielectric constant. Therefore, if the transition dipole moment is known, one extracts the exciton density n_{1s}. The dipole moment for the Lyman $1s - 2p$ transition is calculated by direct transposition of the hydrogen atom case, as discussed in Ref. [13]. The temperature measurement rests on the fact that the transition obeys a Boltzmann line shape reflecting the difference of effective mass of the exciton in the $n = 1$ and $n = 2$ states, as illustrated in Fig. 1.

The experimental scheme is shown in Fig. 2. A naturally grown single crystal of Cu_2O with a (100) plane and a 220 μm thickness is mounted in a liquid He cryostat and kept at 4.2 K. Both the pump pulse used to create $n = 1$ orthoexcitons by two-photon transition and the probe pulse used to measure the $n = 1$ to $n = 2, 3, \ldots$ Lyman series transition are derived from the same regenerative amplified Ti:sapphire laser. By frequency down conversion with OPA systems, one produces tunable pulses of a 100 fs duration

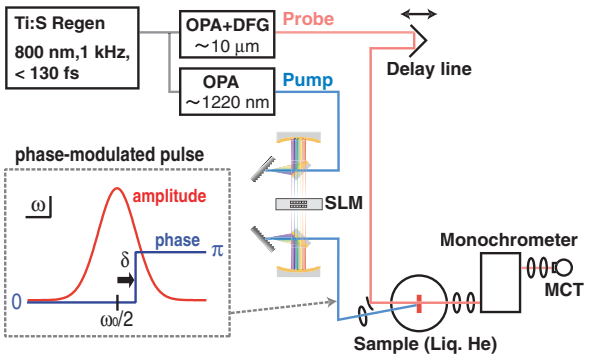

FIG. 2 (color online). Experimental setup.

in the required spectral region (~1.016 eV for the pump; ~116 meV for the probe). The pump pulses are introduced to a pulse shaper in a $4f$ configuration with paired grating and a liquid crystal spatial light modulator (SLM) [1]. We obtain exciton Lyman signals by measuring the differential transmission spectra with and without pump irradiation. Details of the experimental setup and the procedure of Lyman spectroscopy can be found in Refs. [11,13,14].

Figure 3(a) shows the midinfrared absorption spectrum for two different IR laser pump intensities. The $1s$-$2p$ (116 meV) and $1s$-$3p$ (129 meV) terms of the Lyman series are clearly observed at low density [11]. The increase of exciton temperature at high intensity is responsible for the growth of the tail on the higher energy side of the $1s$-$2p$ exciton line. As can be seen in Fig. 3(b), this broad high energy tail grows with I^3, showing that the hot excitons are indeed created in a three-photon process. Another possible mechanism of heating would be the inelastic collision of excitons [7,8]. However, in the present experimental scheme, the production rate of hot carriers through such an Auger process is unlikely since it would be proportional to I^4.

The problem is to reduce this unwanted three-photon contribution without reducing the production rate of cold excitons by two-photon absorption. This can be achieved by coherent control of the multiphoton transitions involved by tailoring the phase and amplitude of pulses into a desired condition as was demonstrated in atomic systems [2,3]. Using a phase-modulated optical pulse having unchanged power spectrum and energy, the two-photon transition rate to the discrete orthoexciton state remains unchanged while the three-photon transition probability to the band-to-band continuum is greatly reduced.

For short pulse two-photon excitation of a two-level system separated by an energy $\hbar\omega_0$, the two-photon transition probability in the absence of near resonant intermediate states can be written as [3]

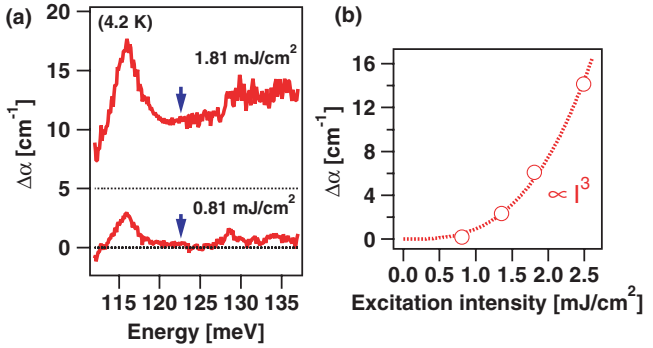

FIG. 3 (color online). (a) Typical differential midinfrared transmission spectra in the region of exciton internal transitions of Lyman series for low (0.81 mJ/cm^2) and high densities (1.81 mJ/cm^2). (b) Pump power dependence of the signal monitored at the high energy tail [123 meV; downward arrows in (a)].

$$P^{2\mathrm{ph}} \propto \left| \int_{-\infty}^{\infty} \varepsilon(\omega_0/2+\Omega)\varepsilon(\omega_0/2-\Omega)d\Omega \right|^2 = \left| \int_{-\infty}^{\infty} A(\omega_0/2+\Omega)A(\omega_0/2-\Omega) \times \exp[i\{\phi(\omega_0/2+\Omega)+\phi(\omega_0/2-\Omega)\}]d\Omega \right|^2, \quad (2)$$

where $\varepsilon(\omega) = A(\omega)\exp[i\phi(\omega)]$ is the electric field of the excitation pulse in frequency domain, and $A(\omega)$ and $\phi(\omega)$ are the spectral amplitude and phase, respectively. The multiphoton transition rate is cast in a form that highlights the superposition of many transition paths with appropriate phase components to the final state. It shows that an excitation pulse of central frequency at $\omega_0/2$ can effectively cycle optimally all photons even if the bandwidth of the laser is much larger than the considered transition. This is immediately apparent for a Fourier transform-limited pulse (for which the spectral phase of the pulse is flat) because each photon with a defect of energy $\omega - \omega_0/2 = \Delta$ can pair with a simultaneous photon of excess energy Δ to fulfill the energy conservation law [15].

In most cases, if one modifies the spectral phase of the pulse, one loses this advantage. For instance if one impresses a linear chirp to the pulse (quadratic phase shift), photons with an energy $\omega_0/2 - \Delta$ have no matching partner of frequency $\omega_0/2 + \Delta$ coincident in time. There are, however, special phase-manipulated pulse shapes, which have the same two-photon transition probability as the Fourier transform-limited pulse but a greatly reduced three-photon transition rate. One example is a pulse having a flat spectral phase with an abrupt π phase shift at $\omega_0/2$.

To demonstrate the property of the π phase shifted pulse in the two-photon process, we monitor the signal at the peak of the $1s$-$2p$ orthoexciton transition energy (116.5 meV) that is proportional to the cold $1s$ orthoexciton density generated by the two-photon excitation as a function of the phase step frequency position [Fig. 4(a)]. The two-photon transition rate is decreased due to destructive interference between the different transitions paths, except in a narrow frequency region around $\Delta \sim 0$,where it reaches the value of a Fourier transform pulse with perfect constructive interference.

Next, we examine the effect of the π phase shifted pulse on the three-photon process. We perform the same experiment except that we set the detection energy to the high energy tail of the $1s$-$2p$ orthoexciton line at 122.5 meV [Fig. 4(b)]. At this probe energy, only hot excitons generated by the three-photon process contribute to the signal. When the step π phase shift position is tuned to the center frequency of the laser pulse ($\delta = 0$), the three-photon transition rate is strongly reduced. This is in agreement with the theory, since the interference effect between transition paths is suppressed for a multiphoton transition with a broad final state distribution [3]. In this case the transition rate depends only on the temporal waveform of the excitation pulse. Because our tailored pulse has a double humped temporal shape with a reduced peak intensity, it significantly reduces the three-photon transition rate.

As we can see from Figs. 4(a) and 4(b), by changing the π phase step position δ, we can easily control the relative contribution of the two types of transitions. At $\delta/\Delta\omega \sim 0$, ($\pi$ phase shifted pulse) the three-photon transition is strongly suppressed and the two-photon transition dominates. At $\delta/\Delta\omega = \pm 0.3$, the two-photon transition is quenched. $\Delta\omega$ is the FWHM bandwidth of the power spectrum of the pulse.

Figure 5 shows the differential absorption spectra at a probe delay of 5 picoseconds under an excitation with a π phase shifted pulse ($\delta/\Delta\omega \sim 0$) and with a transform-limited (TL) pulse. At low density excitation, both spectra contain signals corresponding to cold $1s$ orthoexcitons directly generated via resonant two-photon absorption. At high density excitation, the spectra of the TL pulses reveal the presence of hot excitons, while the spectra with the π phase shifted pulse reveal only cold excitons with a density of 10^{15} cm^{-3}.

Finally, we discuss the optimum conditions to generate and accumulate cold excitons by resonant two-photon transition with tailored pulses. As we have shown, the recipe is to keep a perfect constructive interference of the

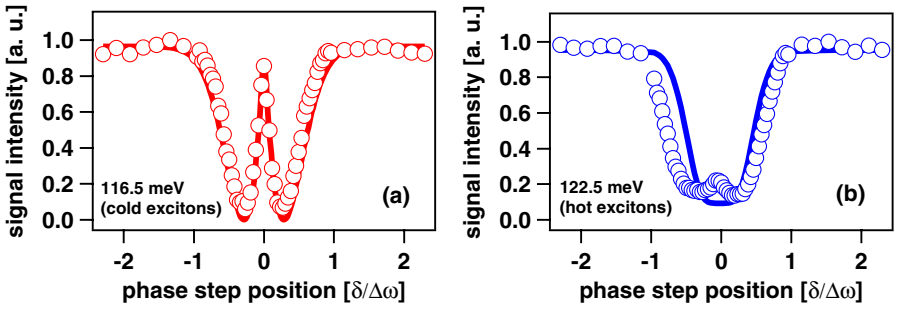

FIG. 4 (color online). Differential absorption signal as a function of the π phase step frequency position for cold excitons (a) monitored at the peak of the $1s$-$2p$ orthoexciton transition energy (116.5 meV) and hot excitons (b) monitored at high energy side (122.5 meV).

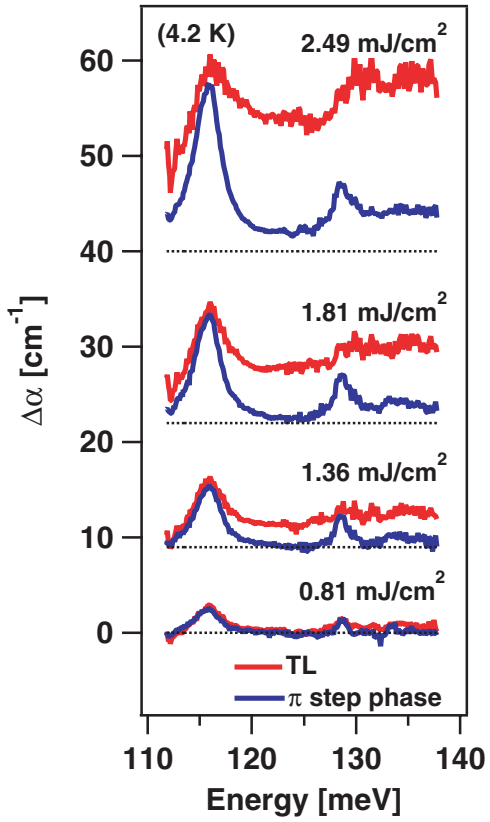

FIG. 5 (color online). Differential absorption spectra of the probe beam at a time delay of 5 picoseconds with a π phase step frequency of $\delta \sim 0$ (lower curves) and with the transform-limited (TL) pulses (upper curves with broad features).

transition paths to optimize two-photon transition but with a decreased peak height of the pulse to suppress the three-photon transition to the band-to-band continuum. For TL pulses, we expect a better contrast for cold excitons using a longer pulse duration. The limit of the pulse duration to obtain perfect interference is determined by the exciton homogeneous linewidth. According to recent high resolution spectroscopy, $n = 1$ orthoexcitons have a linewidth of the order of 1 μeV [16]. From our numerical simulation, the optimum condition for the TL pulse corresponds to a duration of several tens of ps. In practice, however, it is rather difficult to obtain clean manipulated pulses with such a pulse duration. An alternative method to suppress unwanted hot carrier generation is to apply to the TL pulses a sinusoidal phase modulation with a short period.

In conclusion, using phase-manipulated pulses we demonstrated that it is possible to select the desired multi-photon transition in semiconductors. We exploited this coherent control to suppress exciton heating of orthoexcitons in Cu_2O. A supercold exciton gas with a density up to 10^{15} cm^{-3} was obtained at a lattice temperature of 4.2 K. Further careful optimization of the excitation pulse shape and a reduction of the lattice temperature below 1 K could lead to a quasisteady state quantum degenerate exciton gas at a density of 10^{16} cm^{-3}. The conditions for BEC of a dilute Bose gas of excitons would be then realized.

We thank T. Tayagaki and T. Shimasaki for fruitful discussions, K. Misawa and K. Horikoshi for their advice about pulse-shaping technique, and T. Sakamoto for experimental support. We also thank J. B. Héroux for critical reading of the manuscript. This work is partly supported by the Grant-in-Aid for Scientific Research (S) from Japan Society for the Promotion of Science (JSPS), and Special Coordination Funds for Promoting Science and Technology from the Ministry of Education, Sports, Science and Technology of Japan (MEXT).

*gonokami@ap.t.u-tokyo.ac.jp

[1] A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).
[2] D. Meshulach and Y. Silberberg, Nature (London) **396**, 239 (1998).
[3] D. Meshulach and Y. Silberberg, Phys. Rev. A **60**, 1287 (1999).
[4] K. A. Walowicz, I. Pastirk, V. V. Lozovoy, and M. Dantus, J. Phys. Chem. A **106**, 9369 (2002).
[5] V. V. Lozovoy, I. Pastirk, K. A. Walowicz, and M. Dantus, J. Chem. Phys. **118**, 3187 (2003).
[6] A. Mysyrowicz, D. Hulin, and A. Antonetti, Phys. Rev. Lett. **43**, 1123 (1979).
[7] D. W. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. B **41**, 11 171 (1990).
[8] K. E. O'Hara, J. R. Gullingsrud, and J. P. Wolfe, Phys. Rev. B **60**, 10 872 (1999).
[9] D. W. Snoke, J. P. Wolfe, and A. Mysyrowicz, Phys. Rev. Lett. **59**, 827 (1987).
[10] T. Goto, M. Y. Shen, S. Koyama, and T. Yokouchi, Phys. Rev. B **55**, 7609 (1997).
[11] T. Tayagaki, A. Mysyrowicz, and M. Kuwata-Gonokami, Phys. Rev. B **74**, 245127 (2006).
[12] K. Yoshioka and M. Kuwata-Gonokami, Phys. Rev. B **73**, 081202(R) (2006).
[13] T. Tayagaki, A. Mysyrowicz, and M. Kuwata-Gonokami, J. Phys. Soc. Jpn. **74**, 1423 (2005).
[14] M. Kuwata-Gonokami, M. Kubouchi, R. Shimano, and A. Mysyrowicz, J. Phys. Soc. Jpn. **73**, 1065 (2004).
[15] It should be noted that for the case of pulsed two-photon transition to a discrete level, the two-photon transition with sufficiently short pulses does not depend on the pulse duration. Therefore we cannot define the two-photon absorption coefficient β parameter unambiguously. Moreover, the transition probability depends on the phase profile. Therefore, in order to extract a parameter characterizing the two-photon transition efficiency, it is necessary to specify the optical pulses completely (phase and amplitude).
[16] G. Dasbach, D. Fröhlich, H. Stolz, R. Klieber, D. Suter, and M. Bayer, Phys. Rev. Lett. **91**, 107401 (2003).