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Quantum inelastic collisions between paraexcitons in Cu_2O

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A density-dependent loss of optically inactive paraexcitons is measured as a function of temperature over a wide range of densities in Cu_2O. At low temperatures, the two-body inelastic collision rate stays at a constant and the extracted collision cross-section diverges. A loss channel cannot be explained classically but is expected in a quantum-mechanical regime. We discuss the repercussion of this loss on the feasibility of achieving Bose-Einstein condensation of paraexcitons in Cu_2O.

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Recent advances in manipulating atomic gases has led to a deeper understanding of macroscopic coherent quantum states. In particular, a smooth evolution between Bose-Einstein condensation (BEC) and a BCS-type phase has been investigated experimentally.[1,2] Such a progress could be achieved because well-designed cooling and trapping of atoms can eliminate inelastic collision channels in long-lived atomic states so that interatomic interactions at sufficiently low temperature are ruled by a constant elastic-scattering length.

BEC of excitons ("atoms" in semiconductors) and the crossover from a BEC to a BCS-type phase have also attracted much attention both experimentally and theoretically. However, the case of excitons is much less advanced. Exciton-exciton interactions are only partially understood due to the many-body nature of the elementary excitation[3–6] and the added complication of the strong coupling to the radiation field as well as the lattice environment. The experimental evidence of exciton BEC is still controversial.[7–9]

A case in point is Cu_2O, *a priori* a favorable direct-gap material where the lowest lying, tightly bound excitons are long lived. Much work has been devoted in searching a BEC phase of 1*s* orthoexciton (the second lowest energy level) gas with a density of 10^{17} cm^{-3} at 2 K. However, this phase has been shown to be unattainable by quantitative photon counting emission measurements[10–14] that revealed an efficient two-body collision-induced loss process. The origin of the density-dependent decay was first attributed to an Auger recombination effect,[15] a process in which a pair of excitons disappears in each inelastic collision, giving rise to a single pair of hot electron-hole free carriers although a theoretical evaluation reports much smaller loss rates than experiment.[16] Later, a theoretical estimate[17] showed that inelastic collisions between pairs of orthoexcitons leading to a conversion into paraexcitons is more probable than the Auger process. Attention therefore now focuses on 1*s* paraexcitons with a negligible Auger coefficient[16] and we should study their stability at high densities as the prime candidate for exciton BEC.

In this Rapid Communication, we report on a systematic study of an inelastic collision process between 1*s* paraexcitons in Cu_2O at low temperatures. These singly degenerate particles have a pure triplet spin configuration,[18] therefore they are ideal excitons in a sense that they effectively do not couple with the radiation field. This makes the exciton density measurements with luminescence difficult. Direct and quantitative measurements of the paraexciton density can be performed in a steady-state regime by the excitonic Lyman spectroscopy.[19] The long lifetime of paraexcitons enables us to vary and probe their density over a wide range by use of a cw excitation, under complete thermalization with respect to the lattice. Therefore one can measure the density-dependent loss rate at low temperature in an equilibrium condition, which can hardly be obtained by luminescence decay measurements under a pulsed excitation.[14] We have discovered a two-body inelastic collision process with a temperature independent rate at low temperature, indicating a diverging cross-section which cannot be explained by the classical hard-sphere collision model. Such feature is a clear signature of the inelastic collision of quantum-mechanical particles. This process removes excitons and leads to a marked saturation at high densities. Based on the obtained loss rate at low-temperature limit, we discuss the feasibility of BEC of paraexcitons.

The sample is a high-quality, natural single crystal of 220 μm thickness with the [100] crystal axis oriented perpendicular to the sample main surface. It is placed in a dynamic helium flow cryostat. The sample is irradiated with a cw single-mode dye laser set at 601 nm. The absorbed light generates 1*s* orthoexcitons [see the inset of Fig. 1(a)] rather homogeneously along the excitation direction via the LO phonon-assisted absorption with the measured small absorption coefficient of typically $\alpha=34$ cm^{-1} at 20 K. The temperature-dependent absorption coefficient is carefully measured at each temperature in order to predict the number and the density profile of orthoexcitons created. A 70% production efficiency is adopted from Ref. 10 but its precise

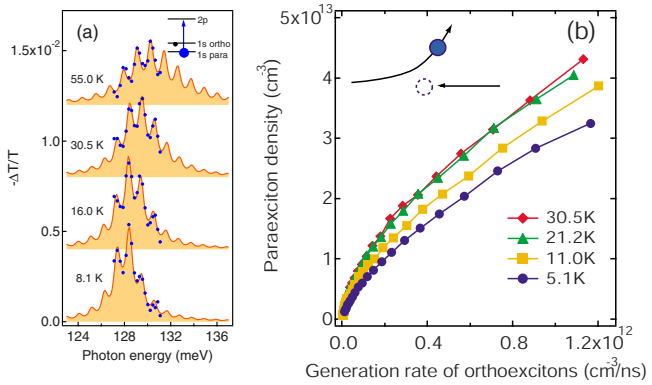

FIG. 1. (Color online) (a) Typical $1s$-$2p$ induced absorption spectra of paraexcitons at various temperatures. The zero levels are offset for clarity. The absorbed excitation density is 7.4 W/cm^2. Solid circles are experimental data and the discrete spectral values correspond to the 9P branch oscillation lines of the CO_2 laser. Filled area is the calculated spectrum taking into account the thermal distribution of the $1s$ state and the bandwidth of the $2p$ state. The oscillatory structure is due to the Fabry-Perot effect on the probe beam inside the sample. (Inset) Schematic of the corresponding transition. The size of the closed circles roughly indicates the population. (b) Paraexciton density at various lattice temperatures as a function of the generation rate of $1s$ orthoexcitons. The generation rate is known from the absorbed power density and the quantum efficiency for creation of orthoexcitons. (Inset) Schematic of the corresponding collision process.

value does not affect our conclusions (see below). It is known that orthoexcitons convert into paraexcitons within a few nanosecond by a spin-flip process in the range of lattice temperatures considered here.[20–22] In view of the lifetimes of orthoexciton and paraexciton, ($\tau_{para} \sim 1$ μs, see below; $\tau_{ortho} \sim 1$ ns) the ratio of paraexciton to orthoexciton under quasi-cw conditions is around 10^3: therefore the exciton population under our cw excitation regime consists essentially of singly degenerate paraexcitons and the influence of orthoexcitons in exciton-exciton interactions can be safely neglected. The mid-infrared probe beam is obtained from a single-line cw CO_2 laser, which can be tuned in discrete steps in the region of interest, covering most of the $1s$-$2p$ line profile. Changes in the transmitted probe beam on the order of 10^{-6} could be detected. A detailed description can be found in Ref. 19. Figure 1(a) shows typical induced absorption spectra taken at various lattice temperatures. The maximum absorbed light intensity is 13 W/cm^2, which corresponds to a orthoexciton generation rate of 1.2×10^{12} cm^{-3}/ns. No intensity-dependent change in spectral profiles is observed, which excludes any heating process due to the excitation. Reconstruction of the entire absorption line profile from the limited spectral range of the data allows extracting the oscillator strength of the transition. Good fits of the data to theoretical absorption curves are obtained by assuming that the temperature of paraexciton gas is the same as the lattice temperatures.

From the induced absorption spectra and the obtained oscillator strengths, we calculate spatially averaged paraexciton densities using the following equation (in SI units):

$$n = \frac{\hbar c \epsilon_0 \sqrt{\epsilon}}{\pi |\mu_{1s\text{-}2p}|^2 E_{1s\text{-}2p}} \int \Delta\alpha_{1s\text{-}2p}(E) dE, \tag{1}$$

where ϵ_0 is the dielectric constant in vacuum, $\sqrt{\epsilon}$ the index of refraction of Cu_2O at around 10 μm, and $E_{1s\text{-}2p}$ the $1s$-$2p$ transition energy (128 meV). We assume the same value $\mu^{para}_{1s\text{-}2p} = \mu^{ortho}_{1s\text{-}2p} = 3.5 \pm 0.3$ eÅ (Ref. 23) for the $1s$-$2p$ transition dipole moment of paraexcitons and orthoexcitons. We note, however, that the binding energy of the paraexciton is larger than that of $1s$ orthoexciton by about 10%. As we will mention later, this point can affect the evaluation of the collision loss rate but will not change our conclusions.

Figure 1(b) shows the paraexciton density as a function of the generation rate of orthoexcitons at various lattice temperatures. A nonlinear saturation of paraexcitons is seen even at a density below 10^{14} cm^{-3} (the corresponding interexcitonic distance is 290 nm, to be compared with the paraexciton Bohr radius of about 0.7 nm). This saturation in a dilute situation where $1s$ paraexcitons dominate the system clearly indicates the presence of a strong paraexciton-paraexciton inelastic collision process responsible for a loss of particles.

In order to extract the collision-induced loss rate from the data, we performed a three-dimensional, explicit numerical modeling of the nonlinear behavior with the following coupled rate equations, where $n_{o,p} = n_{o,p}(\mathbf{r},t)$ is the position-dependent density of orthoexciton (o) and paraexciton (p)

$$\dot{n}_o = D_o \nabla^2 n_o - \frac{1}{\tau_{op}} n_o + \frac{3}{\tau_{op}} e^{-\Delta_{op}/k_B T} n_p - 2A(n_o + n_p) n_o + \frac{3}{4} A (n_o + n_p)^2 + G, \tag{2}$$

$$\dot{n}_p = D_p \nabla^2 n_p + \frac{1}{\tau_{op}} n_o - \frac{3}{\tau_{op}} e^{-\Delta_{op}/k_B T} n_p - 2A(n_o + n_p) n_p + \frac{1}{4} A (n_o + n_p)^2 - \frac{1}{\tau_p} n_p. \tag{3}$$

These equations take into account the spatial inhomogeneity of the excitons and their migration through the sample as well as the temperature-dependent decay time of paraexcitons. D_o and D_p are the temperature-dependent diffusion coefficients of orthoexciton and paraexciton. Reference 24 reports D_p (see Ref. 25), and $D_o = D_p/3$ (Ref. 10) is assumed here. τ_{op} is the fast, temperature-dependent orthoexciton-paraexciton conversion time (see Refs. 14, 21, and 22), $\Delta_{op} = 12$ meV is the splitting between orthoexciton and paraexciton levels. $G = G(\mathbf{r},t)$ is the position-dependent generation rate of $1s$ orthoexcitons taking into account the beam size of the excitation beam and the absorption coefficient α. τ_p is the temperature-dependent lifetime of $1s$ paraexcitons in the low-density limit (shown below). A is the inelastic collision coefficient for paraexcitons. Although the collision rate for orthoexcitons is very small under our moderate cw excitation, for the sake of completeness the same inelastic collision coefficient A was also introduced for orthoexcitons. Finally, the third term of the right-hand side of the equations corresponds to a thermally activated up-conversion process from paraexcitons to orthoexcitons that becomes important at high

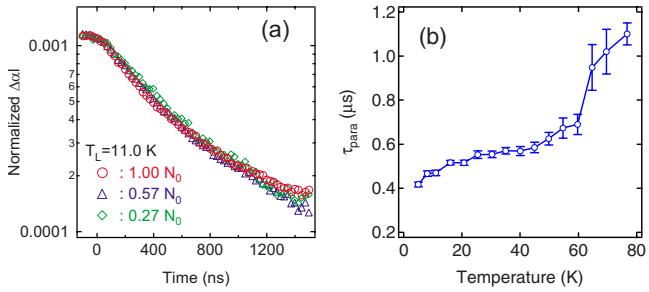

FIG. 2. (Color online) (a) Time-resolved paraexciton population at $T_L=11.0$ K. Absorbed excitation densities are $I_0=0.16$ W/cm^2 (circles), $0.57I_0$ (triangles), and $0.27I_0$ (diamonds). The magnitudes of the signal are normalized at zero time delay. (b) Temperature dependence of the paraexciton lifetime.

temperatures ($T>40$ K). We numerically integrate the equations until the system reaches a steady state and we take the spatially averaged density.[26]

The decay time of paraexcitons τ_p is much shorter than the paraexciton radiative lifetime and can be largely sample dependent. It is also important to introduce the proper low-density value of the decay in the numerical modeling. For this reason, we have also measured the temperature dependence of the paraexciton decay time in our sample at very low densities. Details of this time-resolved method can be found in Ref. 19. In order to ensure a low-density and homogeneous excitation, here we adjusted the excitation wavelength so that the absorption coefficient was $\alpha=5$ cm^{-1}. Figure 2(a) shows a semilog plot of $\Delta\alpha l$ at lattice temperature $T_L=11$ K as a function of time for three excitation intensities. Open circles correspond to an absorbed pump power density of 0.16 W/cm^2 and a generation rate of orthoexcitons of 1.5×10^{10} cm^{-3}/ns. The absence of a dependent decay when reducing the excitation intensity by a factor 2 and 4 ensures that the low-density limit lifetime of paraexcitons is indeed measured. By also taking into account the lateral diffusion, we extract the lifetime of paraexcitons to be 470 ± 12 ns at 11 K. Figure 2(b) shows the temperature dependence of the lifetime of such dilute $1s$ paraexcitons. The reduction in the lifetime below 80 K indicates the presence of capture centers, probably Cu vacancies. The measured decay times are introduced as temperature-dependent parameters in the simulations.

Figure 3(a) shows the results of the numerical calculation for a temperature of 16 K. The inelastic collision coefficient A was parametrically changed from 1×10^{-19} to 1×10^{-14} cm^3/ns in logarithmic steps. As can be seen, a good agreement between the calculation and the experiment is found for $A=4\times10^{-16}$ cm^3/ns. We also performed the calculation assuming a three-body collision process and confirmed that it produces much stronger saturation and does not fit the data. By repeating this best-fit procedure for other temperatures, we obtained the temperature dependence of the inelastic collision coefficient A shown in Fig. 3(b). A striking feature is the fact that the coefficient A is almost independent of temperature with an averaged value $A=3.7\times10^{-16}$ cm^3/ns.

Using the coefficient A, we can estimate the temperature

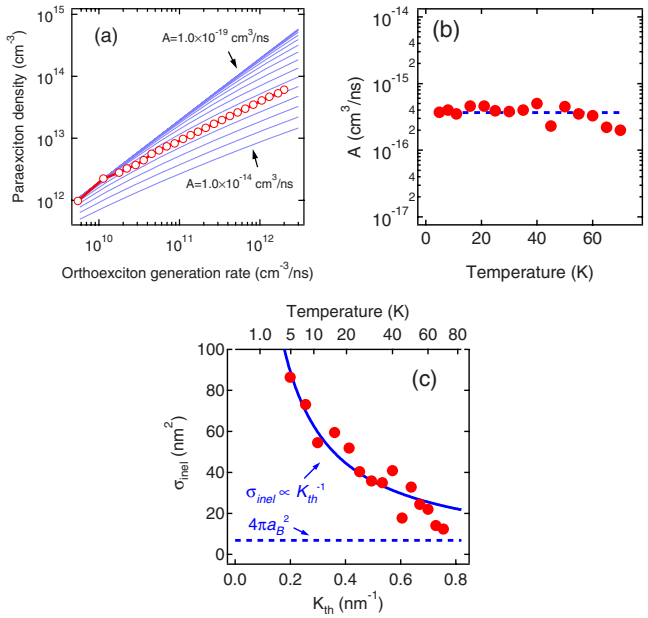

FIG. 3. (Color online) (a) (Solid curves) Calculated paraexciton density as a function of the generation rate of orthoexciton at $T_L=16$ K (see text). (Open circles) Experimental data at $T_L=16$ K. $A=4\times10^{-16}$ cm^3/ns gives the best fit. (b) Temperature dependence of the coefficient A. The coefficient is almost independent of temperature. (c) Exciton-exciton inelastic collision cross-section as a function of the mean thermal wave number of paraexcitons. The dashed curve is a fitted curve assuming $\sigma_{inel}=aK_{th}^{-1}$, where a is a fitting parameter. For comparison, the classical collision cross-section $4\pi a_B^2$ is also shown.

dependence or the momentum dependence of the inelastic collision cross-section σ_{inel}. The total collision rate is $\sigma_{inel}v_{th}n=An$, where $v_{th}=\sqrt{3k_BT/m_{ex}}$ is the mean thermal velocity of excitons (m_{ex} is the effective mass of a $1s$ paraexciton[19,27,28]). As can be seen in Fig. 3(c), a K_{th}^{-1} dependence (or $T^{-1/2}$ dependence) of the inelastic collision cross-section σ_{inel} is revealed as a function of the mean wave number $K_{th}=m_{ex}v_{ex}/\hbar$. This is the evidence of a quantum inelastic collision process: in the fields of nuclear physics and ultracold atoms, it is well known that in the quantum-mechanical limit the inelastic-scattering cross-section at low temperature generally shows a diverging K_{th}^{-1} dependence,[29,30] while the elastic cross-section becomes a constant ($\sigma_{el}=4\pi a^2$, where a is the s-wave scattering length). Quantum effects are expected to be important for excitons in a wide temperature range since an exciton is a very light particle with a comparable effective mass the electron mass at rest. In fact, the temperature at which the thermal de Broglie length becomes comparable to the exciton Bohr radius a_B of the paraexciton is on the order of 10^3 K. This means it is essential to treat the collision processes quantum mechanically at all temperatures. This point applies to exciton systems in any material.

The main uncertainty in evaluating the value $A\sim4\times10^{-16}$ cm^3/ns comes from the adopted values of the $1s$-$2p$ dipole moment and of the quantum efficiency for generating orthoexcitons. For example, if we assume a 50% quantum efficiency instead of 70%, a dipole moment of 3 eÅ instead

of 3.5 eÅ, the coefficient A is reduced by a factor 2. A dipole moment of 3 eÅ is obtained by adopting a Bohr radius of 6.8 Å for the paraexciton taking into account its larger binding energy (150 meV instead of 138 meV for the orthoexciton). Considering all possible errors, we estimate an uncertainty for the value of A to stay within a factor of 3.

A clarification for the surprisingly large "Auger-type" collision coefficient of paraexcitons and the identification of the final products emanating from the collisions remain a challenging task.[14] We note that the coefficient for orthoexcitons have been shown to be similar values.[10–14] We now consider the impact of the temperature-independent collision rate on the feasibility of paraexciton BEC. Considering an exciton thermalization time on the order of a nanosecond, the nonlinear saturation makes it difficult to reach the phase transition at 2 K with the critical density of 10^{17} cm^{-3}. However, at lower temperatures, where the critical densities for the phase transition are on the order of (10^{14}–10^{16} cm^{-3}), the inelastic scattering becomes insignificant, making the condensation possible. We intend to cool the sample with a ^{3}He cryostat or a dilution refrigerator and trap the gas of excitons inside the crystal to induce this phase transition. A very high-purity sample and the enhancement of paraexciton-TA phonon interactions by applying strain fields[24] would be beneficial for cooling the exciton gas below 100 mK.

In conclusion, we have observed a quantum-mechanical inelastic collision process of optically inactive 1s paraexcitons in Cu_2O. A characteristic, temperature-independent inelastic collision rate leading a strong divergence of the collision cross-section was revealed using the excitonic Lyman spectroscopy. The microscopic mechanism of the large cross-sections of paraexcitons and the final products of the paraexciton collision are open to be made clear. A rigorous treatment of the many-body correlation in the electron-hole system could provide a clue to solve the problem.

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[26] See supplementary material at http://link.aps.org/supplemental/10.1103/PhysRevB.82.041201 for typical results.

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