Relaxation explosion of a quantum degenerate exciton gas in Cu$_2$O

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Abstract. We present our recent experimental studies on anomalous luminescence and its connection to Bose–Einstein condensation (BEC) transition of dark excitons in a bulk semiconductor. Our sensitive and quantitative detection of this nonluminous quasi-particle using hydrogen-like internal transitions allows obtaining continuous spectra of dark excitons using a quantum cascade laser. According to quantitative measurements on the two-body inelastic collision cross section of excitons, the system needs to be cooled to sub-Kelvin temperatures. We discuss in detail our recent observation of an explosive phenomenon when the BEC criterion is satisfied (Yoshioka et al 2011 Nature Commun. 2 328) for trapped excitons using a helium-3 refrigerator, and outline a plausible scenario when the BEC transition occurs in an inelastic environment. We also discuss how to increase the condensate fraction in order to study the unique ground state of many-body electric excitations in solids.
1. Introduction

It has been almost 50 years since the Bose–Einstein condensation (BEC) of excitons was first proposed theoretically as a possible candidate to demonstrate BEC [1–3]. With the realization of BEC in dilute atomic gases leading to a clear understanding of the macroscopic quantum state of weakly interacting bosons, there is growing interest in elucidating the nature of the ground states of the quasi-particles in many-body electrons in semiconductors. Thus, it is pivotal to investigate how the finite non-bosonic contribution of the composite particles results in novel collective phenomena of the many-body exciton system, as well as the observation of the BEC transition itself. For example, the occurrence of superfluidity in the exciton condensate is a long-term problem [3–5]. Until now, however, there have been no clear experimental results to resolve such fundamental issues as introduced in the following.

The 1s para-excitons in direct semiconductor cuprous oxide (Cu$_2$O) are isolated from the radiation field, and thus their lifetime reaches microseconds [6, 7]; this allows us to treat the exciton gas as a purely matter-like system under continuous supply and loss of excitons. In addition, it provides a unique opportunity to investigate the spontaneous quantum phase transition of the matter waves as a nonequilibrium and open system, without using the coherence of photons as in the case of cavity exciton–polaritons [8–10] and photons in a microcavity [11]. The search for BEC of indirect excitons in coupled quantum well structures has been conducted similarly [12]. However, the decoupling from the electromagnetic field in turn prevents reliable measurements of the density and temperature of para-excitons through conventional experimental techniques, such as lineshape analysis of the exciton luminescence spectrum [13]. Thus, it is difficult to evaluate the parameters of para-excitons that are crucial to clarify the BEC transition, i.e. effective mass, lifetime and most importantly the two-body collision-induced loss rate. The quantitative measurement of the collision-induced loss rate for luminous 1s ortho-excitons has cast serious doubts on the quantum statistical lineshape in the luminescence spectra [14]. The measured inelastic collision coefficient is several orders of magnitude larger than the theoretical estimation based on exciton Auger processes [15]. In this sense, the origin of the two-body loss is still unclear, and the magnitude of two-body loss for para-excitons has become a critical issue. Peculiar luminescence spectra [13] and ballistic photovoltaic signals [16] that imply the occurrence of para-exciton BEC and its superfluidity have been reported, but the estimated densities were uncertain since the inelastic scattering loss coefficient was unknown. Therefore, the development of experimental techniques other than luminescence spectroscopy is necessary to quantitatively evaluate the density and temperature of dark para-excitons.

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In this study, we first review our technique that achieves sensitive detection of the dark para-excitons using a laser-based spectroscopy. We present our recent results on the detection of steady-state para-excitons using a mid-infrared quantum cascade laser (QCL). Based on the measured two-body collision-induced para-exciton decay, we briefly explain the reason for cooling the exciton for BEC to sub-Kelvin temperatures. Next, we present detailed experimental data of our recent publication on the indirect evidence for the transition to an exciton BEC in Cu$_2$O, observed using a helium-3 refrigerator [17]. Finally, we discuss future prospects of realizing pure and macroscopic exciton condensate by cooling the system to the sub-100 mK region.

2. Excitonic Lyman spectroscopy of steady-state para-excitons

The exciton system in Cu$_2$O exhibits well-separated hydrogen-like energy levels, and therefore it is possible to apply fine laser spectroscopy that is usually performed in atomic physics. We have used the hydrogen-like 1s to np ($n = 2, 3, 4, \ldots$) absorption measurements that appear only when 1s excitons are present. The resonances are in the mid-infrared region (typically 10 µm). This so-called excitonic Lyman spectroscopy is capable of unambiguously determining not only the density of dark excitons but also their thermal distribution functions. The latter is due to the difference in the effective masses of 1s and np excitons. The origin of the mass difference can be explained as follows. The spatial spread of the np excitons extends over tens of unit cells, and thus the Wannier–Mott exciton picture perfectly describes the hydrogen-like relative electron–hole motion with a Rydberg energy of 98 meV. The effective masses in these cases are simply the sum of the effective masses of the electron and the hole at band minima, $1.68m_0$ ($m_0$ is the electron mass at rest). On the other hand, 1s excitons are tightly bound electron–hole pairs with a Rydberg energy of 151 meV and their radii are comparable to the lattice constant. Therefore, their electron and hole wavefunctions in $k$-space extend to nearly half of the first Brillouin zone. This results in a greater effective mass of the constituent particles, and therefore, the effective mass of a 1s para-exciton is $M_{ex} = 2.6m_0$ [18, 19], which is heavier than np excitons.

Assuming vertical transition (i.e. no change in exciton momentum in the transition) of the 1s–np absorption, the following exciton wavenumber ($K$) dependence of the transition energy can be deduced from the energy conservation rules:

$$E_{np}(K) - E_{1s}(K) = E_{np}(0) - E_{1s}(0) + \left( \frac{1}{m_{np}} - \frac{1}{m_{1s}} \right) \frac{\hbar^2 K^2}{2}.$$  

(1)

Therefore, the 1s–np transition energy is momentum dependent because of the difference in the effective masses of the initial and final states, as described above. In addition, the 1s ortho-exciton and 1s para-exciton levels are split by 12 meV because of the electron–hole exchange interactions, while the np levels are degenerate. In most cases, we use 1s–2p-induced transitions to separately detect both 1s ortho-excitons and 1s para-excitons within the spectral transmission window of the background material. Although the final state exhibits a broad linewidth (1 meV) that hinders the evaluation of the exciton temperature below 10 K, this method still sensitively and quantitatively detects the density of 1s excitons at any temperature. Ultrafast conversion from the 1s ortho-exciton to 1s para-exciton level was investigated by mid-infrared femtosecond pump–probe spectroscopy [20–22]. A non-perturbative para-exciton response
under a strong mid-infrared pulse resonant with 1s–2p transition was also observed [23].

Terahertz spectroscopy was applied to other internal transitions such as 3p–2s [24]. Continuous-wave (cw) excitonic Lyman spectroscopy is suitable [18, 25, 26] for a systematic study of long-lifetime 1s para-excitons. Unlike 1s ortho-excitons that have nanoseconds lifetimes, 1s para-excitons can be accumulated to high density under a cw excitation, allowing us to study nonlinear phenomena such as two-body inelastic collisions in a steady-state regime. A wide dynamic range of five orders of magnitude was demonstrated by sensitively measuring the induced absorption by using a cw, line-selective carbon dioxide laser [18]. The carbon dioxide laser is a good mid-infrared probe source when it is used to investigate relatively slow dynamics such as the lifetime of para-excitons by using one oscillation line only. However, only a sparse spectrum can be obtained because of the limited number of oscillation wavelengths (i.e. 9P branch lines). In this study, we developed a setup using a continuously tunable QCL with an external cavity (Daylight Solutions, Model 10096-010-M532J) as an alternative probe light source. The smallest changes in the induced absorption spectra, originating from exciton–exciton interactions or occurrence of the para-exciton BEC, would be detected if a sensitive technique to detect continuous spectra is developed.

Figure 1 depicts the experimental setup. An acousto-optic modulator generates rectangular pulses from the cw pump light of a ring dye laser (Coherent, Inc., 899-21). The probe QCL operates in a pulsed mode. The durations of the pump and probe pulses are 6000 and 120 ns, and the repetition rates are 5 and 10 kHz, respectively. The relative delay of the probe was set immediately before the end of the pump pulse so that the probe detects steady-state para-exciton gas. The pulse energy of the pump was 700 nJ. We attached an aperture on the sample to ensure spatial overlap between the pump and the probe, which are collimated and not focused on the sample. A cooled mercury cadmium telluride (MCT) photodiode with a preamplifier detects the mid-infrared probe pulses. A 16-bit, fast A/D converter records every probe signal that is used to calculate the differential transmission. A dynamic helium flow cryostat cools the sample.

Although the QCL has an external cavity, the oscillation wavelength of the probe laser hops within a few neighboring longitudinal modes on a shot-to-shot basis. Although the intensity of the probe laser pulse is stable, we found that the probe pulse intensity detected by the MCT detector fluctuates in a quantized manner (see figure 2). This is because the cryostat windows and the Cu$_2$O crystal as a whole behave as a Fabry–Pérot interferometer because of their internal reflections. Namely, the transmittance of the optical elements is wavelength dependent. Hence, the pulse-to-pulse mode hopping results in severe intensity fluctuation when detected after transmitting all the optical elements. Therefore, noise occurs when the differential transmission is calculated by merely averaging many probe signal pairs. To resolve this issue, we categorized the detected probe signals by their pulse heights and calculated the differential transmission. We can judge that neighboring two pulses having the same wavelength arrive if their signal heights are almost the same. Therefore, we selected only probe pairs with the neighboring probe pulses having the same wavelengths (nearly equal pulse heights) and calculated the differential transmission. In addition, we used a 5.3 mm thick natural single crystal and set a relatively long absorption length ($1/\alpha = 2$ mm) to obtain a large induced absorption. The thick crystal eliminates the spectral fringe in the induced absorption spectra observed in our previous results using 0.22 mm thick crystals [18].

The right panel of figure 2 shows the improved results for the 1s–2p-induced absorption of para-excitons. Although the minimum detectable transmission change ($10^{-4}$) is an order of magnitude lower than our previous result using a carbon dioxide laser, we can now obtain
Figure 1. Experimental setup for cw excitonic Lyman spectroscopy of para-excitons in cuprous oxide using a pulsed QCL. The QCL operates at 10 kHz. We multiplied this frequency by 1000 to synchronize the QCL with a function generator (5 kHz) that was used for chopping the pump laser. Collimated pump and probe beams are sent onto the 5.3 mm thick crystal. We attached an aperture with 1 mm diameter on the incident surface of the sample. An MCT photodiode detects the probe pulses and an A/D converter records the signal synchronously.

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continuous 1s–2p spectra of dilute para-excitons. The spectral areas of the induced absorption are about 1 cm$^{-1}$ meV, corresponding to the density of about $10^{14}$ cm$^{-3}$, confirming that the system sits in a completely classical regime (the BEC critical density at 5 K is $4 \times 10^{17}$ cm$^{-3}$). We can observe that the low-energy tail of the induced absorption is stronger than expected for the Maxwell–Boltzmann distribution of 1s para-excitons, especially at high temperatures (see 70 K in figure 2). This can be confirmed in the induced absorption spectra taken in the entire temperature range, as shown in figure 3.

This phenomenon does not result from quantum statistical effects since the excitation density is low, but from the higher-energy tail of the 1s–2p absorption spectrum of ortho-excitons. The ortho-exciton-to-para-exciton number ratio (in very weak excitation cases) is determined by the Boltzmann factor,

$$\frac{N_o}{N_p} = 3 \exp\left(-\frac{\Delta}{k_B T}\right),$$

where $\Delta = 12$ meV is the ortho–para splitting, and the factor of three originates from the degeneracy of 1s ortho-excitons. Therefore, ortho-excitons are thermally excited from

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para-excitons, and thus cannot be neglected at high temperatures, typically at $T \geq 40$ K. Figure 4 shows the extracted ortho-exciton-to-para-exciton ratio as a function of temperature. Although the thermal activation model quantitatively explains the result, ortho-exciton signals are still two to three times larger. We believe this is due to the inhomogeneous spatial distribution of para-excitons. Because the ortho-excitons are very dilute, they behave linearly at any position in our excitation condition. On the other hand, para-excitons accumulate to relatively high densities and suffer from the two-body inelastic collision processes, especially near the incident surface of the sample. At low temperatures, 1s–2p ortho-exciton absorption narrows so that its high-energy tail cannot contribute to our spectral region of interest; however, we can still find some remnant signals, especially at the lowest temperatures. The lateral diffusion of long-lived para-excitons becomes prominent at low temperatures [27], and the ortho-exciton-to-para-exciton conversion slows down [28]. These effects possibly make $N_o/N_p$ much larger than the thermal activation model, but we need a wider spectral probe for a definitive conclusion. Another possibility is that there might be small but finite asymmetry in the lineshape of the 2p para-exciton state. The contribution of the 1s–3p absorption of ortho-excitons in the 1s–2p para-exciton region is neglected here, since its spectral area is four times smaller than that of 1s–2p ortho-exciton absorption (therefore an order of magnitude smaller than the 1s–2p para-exciton absorption), and the transition energy is 0.3 meV higher than the 1s–2p para-exciton transition.

Figure 2. (Left) Typical probe intensity taken by an MCT photodiode (not cooled, no preamplifier). The repetition rate of the probe pulse is 10 kHz, and each shot was recorded synchronously by an A/D converter. (Right) Typical 1s–2p para-exciton spectra taken using a tunable, pulsed QCL as a probe light source. The thickness of the sample is 5.3 mm. Green dotted curves are calculated spectra assuming Maxwell–Boltzmann distributions for para-excitons only, with the same exciton temperatures as those of the lattice. Note that the signal deviates from the theoretical curve in the lower-energy tail at high temperatures (see text).
Figure 3. 1s–2p-induced absorption spectra of para-excitons as a function of the lattice temperature. Solid curves are theoretically fitted to the data, taking into account the 1s–2p-induced absorption of ortho-excitons, where the total ortho-exciton-to-para-exciton number ratio is an adjustable parameter. The theoretical curve cannot reproduce the data at low temperatures since the high-energy tail of ortho-excitons is not available. The green dotted curves are calculated spectra taking into account the Maxwell–Boltzmann distributions for para-excitons only. The ortho-exciton and para-exciton temperatures are set to the same value as the lattice temperature.

In our previous studies, we extracted the two-body collision-induced loss of the para-excitons by the quantitative measurements of the para-exciton density as a function of the excitation rate. The temperature dependence of the inelastic cross section shows divergence at low temperatures: the $1/\nu$ law typical of $s$-wave inelastic scattering (or a temperature-independent collision loss rate at a constant density) [29]. Although this temperature dependence is a well-known fact, the most unusual feature is that the obtained cross sections are several orders of magnitude larger than the theoretically estimated ones, which are based on the standard exciton Auger recombination model [15]. In particular, the two-body collision-induced loss coefficient is $A \sim 10^{-16}$ cm$^3$ ns$^{-1}$ (the corresponding inelastic cross section is $\sigma_{\text{inel}} = 85 \text{ nm}^2$ at 5 K, with the error of a factor of three); therefore, the effective para-exciton lifetime $\tau_{\text{eff}}$, as determined by $n/\tau_{\text{eff}} = An^2$, becomes about 100 ps. Considering the exciton–phonon relaxation time of approximately 1 ns, this result suggests that the realization of BEC is impossible at 2 K.
Figure 4. Temperature dependence of the ortho-exciton-to-para-exciton ratio extracted from the lineshape analyses of the 1s–2p spectra. The blue dotted curve is the theoretical ratio assuming the Boltzmann distribution between the two 1s exciton species.

because the effective para-exciton lifetime is very short even if we could prepare the critical density of $10^{17} \text{ cm}^{-3}$.

We have shown that it is now possible to detect the density and thermal distribution of dark para-excitons with sufficient sensitivity by using a continuously tunable QCL or a carbon dioxide laser. This will enable us to measure the smallest spectral change induced by unknown properties of quantum degenerate excitons. To further improve the signal-to-noise ratio, a cw QCL with an external cavity will be an ideal probe source. We expect to expand the dynamic range by an order of magnitude. These experiments are in progress.

We have realized that there remain some errors in the estimated 1s–2p transition dipole moment ($\mu_{1s-2p} = 3.5 \pm 0.3 \text{ eÅ}$ [7]). This causes ambiguity in evaluating the density, and thus, the collision loss coefficient of 1s para-excitons. These quantities are nonlinearly dependent on the dipole moment; therefore, greater precision is imposed on the dipole moment than that on any other parameter discussed here. Reduction of the error by a factor of five or better than the current evaluation should suffice for reliable discussions on the quantum degenerate para-excitons using the elastic and inelastic cross sections. In this regard, a stable excitation pulse source at 1220 nm, possibly an optical parametric amplifier (OPA) pumped by a cw regenerative amplifier, is most suitable. As a probe light source, the difference frequency generation of the OPA will be a solution.

3. Cooling excitons to sub-Kelvin temperatures as a prerequisite to reach the Bose–Einstein condensation phase boundary

As discussed in the previous section, we have developed cw laser-based excitonic Lyman spectroscopy for optically dark excitons in Cu$_2$O. The ability of the excitonic Lyman spectroscopy to sensitively detect the density and temperature of para-excitons enabled us to quantitatively measure two-body (inter-para-exciton) inelastic scattering cross sections. We confirmed that the inelastic cross section diverges at low temperature, which clearly demonstrates the s-wave nature of the scattering [7]. Based on the relatively large
scattering-induced loss, we can conclude that it is impossible to cool the para-excitons to a superfluid helium temperature even if we could realize the critical density at this temperature \((10^{17} \text{ cm}^{-3})\). At this density, the effective lifetime is drastically reduced to about 100 ps, which is an order of magnitude shorter than the time required for para-excitons to reach thermal equilibrium with the lattice. Therefore, setting a much lower critical density is an indispensable prerequisite to reach the phase boundary. Following the density–temperature relationship of the BEC phase boundary, \(k_B T_c = (n/2.612)^{2/3} \times 2\pi h^2 / M_{ex}\), we aim to cool the excitons to sub-Kelvin temperatures by using a helium-3 refrigerator. It is normally impossible to achieve BEC in trapped atomic gases when the inelastic cross section is comparable to the elastic cross section, mainly because of the difficulty of momentum re-distribution in the evaporative cooling process. Thus, the BEC transition in the strong inelastic environment offers a unique system to study.

Due to the limited thermal capacity at low temperatures, minimal heat load needs to be applied to maintain the low temperature and long hold time of the single-shot \(^3\)He refrigerator. In our experiments, the typical hold time is 6 h under irradiation with the excitation laser power of 250 \(\mu\)W. On the other hand, the ballistic diffusion of para-excitons at low temperature prevents the accumulation of excitons to the required density within the limited excitation power. Here, we applied an inhomogeneous strain to the sample crystal (see figure 5) and thus prepared a 3D harmonic trap for para-excitons \([30, 31]\). This configuration benefits from the spatial compression of the exciton gas in a harmonic trap at low temperature, and therefore the critical exciton number \(N_c\) for BEC is proportional to \(T^3\),

\[
N_c = 1.2 \left( \frac{k_B T}{\hbar \tilde{\omega}} \right)^3 ,
\]

while in free space it is proportional to \(T^{1.5}\) (\(\tilde{\omega}\) is the geometrical average of our 3D trap frequencies, \(2\pi \times 19\) MHz). Therefore, it is possible to attain the BEC condition under a quasi-cw excitation using a cw ring dye laser and an acousto-optic modulator as a fast light chopper.

**Figure 5.** Setup of a three-dimensional (3D) harmonic trap for para-excitons. A lens with proper curvature is pressed against the single-crystal sample. The applied force is estimated from the spring constant of a hard spring and the amount of its compression. The magnified imaging section shows the active compensation of the vibration of the cryogen-free helium-3 refrigerator.
Para-excitons are converted from photo-excited ortho-excitons under the irradiation of quasi-cw excitation light, and their number can be varied simply by changing the excitation power. The radiative decay of para-excitons becomes weakly quadrupole allowed under the strain field. We monitor the spatially resolved luminescence spectra of this direct luminescence that is emitted in the radiative recombination process. All the data were taken by an electron-multiplying charge-coupled device (EMCCD) with long-time exposure and averaging. As shown in figure 5, the weak luminescence of the trapped para-exciton cloud is magnified onto the entrance slit of the spectrometer with an EMCCD attached to observe the spatially resolved luminescence spectra. We employed an anti-vibration technique using a position-sensitive diode (PSD) and a pair of piezo-electric transducers (PZTs) to minimize the blur of the luminescence images.

The para-exciton temperature determines the spatial and spectral spread of the gas in our 3D harmonic trap. We have confirmed that the temperature of sufficiently dilute para-excitons is 0.8 K when the para-exciton lifetime in the trap is about 300 ns [17]. When we increase the number of para-excitons to and above the critical number at this temperature, \(7.8 \times 10^8\) excitons, we observed an abrupt increase of a high-energy component; this is in contrast with the ideal case where the macroscopic number of particles occupies the ground state of which the spatial size is determined by the harmonic trap frequencies. In figure 6, we show the specific series of spatially resolved luminescence spectra. The threshold-like behavior also appears when we decrease the lattice temperature while we fix the created para-exciton number to about \(2 \times 10^9\).

In the present case, the para-exciton–para-exciton scattering is highly inelastic, and the elastic-scattering cross section is comparable with, or probably smaller than, the inelastic scattering cross section. This prevents volume expansion by the mean-field energy (see below) of the condensate, and thus, the condensate suffers from the enhanced inelastic scattering because of its small volume arising from the steep trap potential. The full-width at half-maximum (FWHM) of the ground-state wave function \(|\phi_0|^2\) is \(u_0 = \sqrt{\hbar/M_0 \omega_i}\), which is about 1 \(\mu\text{m}\) on average. Therefore, it is decreased by \(1.5 \times 10^{-5}\) in volume compared with the trapped, classical gas in the dilute limit. This is the cause of the ‘relaxation explosion’ in the condensate, which is also the case in BEC in atomic hydrogen [32], and the consequent appearance of the hot exciton component. Rate-equation-based analyses and Monte–Carlo simulation results suggest a condensate fraction ranging from 0.1 to 1% [17]. A distinct feature of this system is that the remnant para-exciton in the inelastic scattering process (the other para-exciton recombines nonradiatively) is always a high-strain-field seeker and is cooled by the cold phonon bath. Therefore, the fast para-excitons mostly return to the bottom part of the trap as cold excitons, even after the inelastic collision processes. In addition, the high-energy para-excitons produced in the explosion rarely collide with the accumulated cold excitons around the bottom of the trap, because of the relatively small elastic cross section.

Let us examine the mean-field expansion of the condensate discussed above. Although the (elastic) scattering length is experimentally unknown, here we adopt a theoretical estimation based on a quantum Monte-Carlo method, \(a = 2.1 a_B\) [33], to roughly estimate the effect. Neglecting the kinetic energy in the Gross–Pitaevskii equation (Thomas–Fermi approximation) and assuming 1% condensate fraction, the spatial spread at FWHM in the \(i\) (\(i = x, y, z\)) direction is

\[
d_i = \frac{1.72}{\sqrt{2} \omega_i} \left( \frac{N a \hbar^2 \omega_i}{M_0^2} \right)^\frac{1}{4},
\]

\(\text{(4)}\)
which in our case is $d_{x,y} = 7.5 \mu m$, and $d_z = 5.9 \mu m$ and therefore, the inelastic scattering loss is decreased by a factor of 100. If this is the case, the bosonic scattering rate into the condensate and the scattering-induced loss rate should be moderate compared with the small-sized condensate without taking into account the mean-field effect. To determine the condensate fraction, taking into account the mean-field effects, numerical simulations such as the Monte-Carlo method will be beneficial for determining the fraction and volume of the condensate self-consistently.

We realized that the condensate cannot be observed in the direct luminescence spectrum, since only para-excitons that satisfy the following energy and momentum selection rules can recombine radiatively,

$$E_0 + \frac{\hbar^2 K_{ex}^2}{2M_{ex}} = \frac{\hbar ck_p}{n_i},$$  

$$\hbar K_{ex} = \hbar k_p,$$  

where $\hbar K_{ex}$ is $\hbar k_p$.
where $E_0$ is the exciton energy at zero momentum, $K_{ex}$ and $k_p$ are wavenumbers of excitons and photons, and $n_i$ is the index of refraction. Therefore, the condensate with zero mean momentum cannot be observed through the direct luminescence signal. In other words, no direct luminescence signal will be detected once we have a pure BEC. Momentum-independent $\Gamma_5^\text{ph}$-phonon-assisted luminescence, which is very weak, or the excitonic Lyman spectroscopy shown here will allow us to access the condensate. To uncover the fluid properties of the condensate, we believe the excitonic Lyman spectroscopy will be a powerful tool because of its sensitivity and capability of detecting ultrafast dynamics. Although its spectral resolution is limited to the linewidth (about 1 meV) of the final state, i.e. 2p para-excitons, fewer problems will arise in investigating the fluid dynamics when nearly 100% condensate fraction can be confirmed by other means such as phonon-assisted luminescence.

To obtain a pure Bose–Einstein condensate (condensate fraction of nearly unity) and to study the properties of this unique many-body ground state of elementary excitations, we need to pursue a much more dilute condensate to avoid an explosion. This can be accomplished by cooling the crystal using a dilution refrigerator and thus lowering the critical temperature by an order of magnitude or more. This not only prolongs the effective lifetime of the para-exciton gas at the critical density but also reduces the heating of the crystal because the excitation power can be lowered nonlinearly. Although we cannot estimate the condensate fraction without knowing the bosonic scattering rate, the BEC transition in lower density should increase the fraction. Because of the frozen interaction between the cold para-exciton gas and the lattice phonons, the para-exciton lifetime in the dilute limit needs to be very long to lower the exciton temperature to 100 mK. This means that a very-high-purity crystal is preferred, since the lifetime is limited by crystal imperfection (the radiative lifetime of para-excitons is estimated to be more than 100 $\mu$s). The exciton–phonon scattering time (and the mobility) is dependent on temperature as $\tau \sim T^{-5/2} - \tau \sim T^{-3/2}$ depending on the applied stress [27]. Considering that the temperature of our para-exciton gas is 800 mK with the lifetime of 300 ns, we estimate that long lifetimes from 5 to 50 $\mu$s are required to reach 100 mK. Our experiments are currently in progress using a selected natural single crystal at a base temperature below 40 mK. Another group recently reported a detailed investigation on high-density, trapped para-excitons using a dilution refrigerator [34]. We envision that we can maintain the density of the condensate low by decreasing the trap frequency and therefore making the volume of the ground state larger. A condensate fraction of typically 10% should be sufficient to be observed as a distinct peak in the weak and noisy signal of longitudinal optical (LO)-phonon-assisted luminescence spectra.

4. Conclusion

We developed sensitive excitonic Lyman spectroscopy for detecting dark 1s para-excitons in Cu$_2$O using quasi-cw mid-infrared lasers. We obtained continuous spectra by using a recently developed QCL. This spectroscopic technique has allowed quantitative evaluation of important parameters for para-excitons. In particular, we found that two-body collision-induced loss of para-excitons, the origin of which is still unclear, prevents BEC from being achieved at a superfluid helium temperature because of the short effective exciton lifetime. We were therefore prompted to cool the excitons to sub-Kelvin temperature using a helium-3 refrigerator, in order to lower the critical density. The trapped para-exciton gas at 0.8 K shows an abrupt emergence of hot excitons above the critical number for ideal bosons. This is attributed to
the relaxation explosion of the condensate, originating from the small condensate volume in the trap potential and the rather large inelastic collision rate between para-excitons. A deeper cooling and lowering of the trap frequency will suppress the explosion of the exciton condensate and produce a significantly higher condensate fraction than the current 0.1–1%. This will enable us to examine the properties of the ground state of matter-like, photo-excited quasi-particles in this bulk semiconductor.

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