Dynamics of excitons in a potential trap at ultra-low temperatures: paraexcitons in Cu$_2$O

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Abstract. We present experiments on the luminescence of excitons confined in a potential trap at sub-Kelvin temperatures after nanosecond pulsed laser excitation. Analysis of the experimental results with a rate model shows that the so-called Auger decay of yellow excitons, which in previous studies led to a rapid decay of the excitons at high densities and thus prevented reaching the critical density for Bose–Einstein condensation (BEC), is greatly reduced for paraexcitons. We demonstrate that exciton numbers well above $10^{10}$ can be collected in a potential trap, albeit at temperatures in the 10 K range. During their lifetime of about 500 ns the paraexcitons cool down to the temperature of the He bath. This opens up the possibility to observe a BEC of paraexcitons provided that the bath temperature can be reduced to below 100 mK.

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1. Introduction

Almost 50 years ago, excitons [1, 2] were suggested as particularly interesting candidates for Bose–Einstein condensation (BEC), as they consist of an electron and a hole in a semiconductor, both fermions bound to form a bosonic excitation and thus resembling most closely atoms of usual matter. Due to their rather small mass comparable to the free electron mass, it was speculated that for exciton densities of the order of $10^{18} \text{cm}^{-3}$, easily achievable by the absorption of photons, critical temperatures of about 10 K may be reached.

As in all physical systems for which BEC has been demonstrated until now, excitons should be confined in a potential trap. This has the advantage that (i) the diffusion process, which reduces the exciton density, is suppressed and (ii) the critical number of particles required for the phase transition [3],

$$N_C = \zeta(3) \left( \frac{k_B T}{\hbar \Omega_0} \right)^3 \tag{1}$$

with $\Omega_0$ being the average oscillator frequency of the trapping potential and $\zeta$ the Riemann zeta function, decreases much faster with temperature, $T$, than in free space.

Due to their unique properties, the excitons of the so-called yellow series in the semiconductor cuprous oxide (Cu$_2$O) are still considered one of several promising candidates for excitonic BEC (for reviews see [4–6]). This is related to the large binding energy of 150 meV, which shifts the Mott density to $10^{19} \text{cm}^{-3}$ at cryogenic temperatures [7, 8]. Made up of doubly degenerate valence and conduction bands, the ground state of this series splits into the triply degenerate orthoexciton and the nondegenerate paraexciton, which is the energetically lowest exciton state, $\Delta = 12.1 \text{meV}$ below the orthoexciton states. Due to the positive parity of the bands, the orthoexciton is only weakly optically allowed (quadrupole transition with oscillator...
strength $3 \times 10^{-9}$ [9]), while the paraexciton as the pure spin triplet state is optically forbidden in all orders. Its decay is only possible via an odd parity optical phonon, resulting in a long lifetime in the microseconds range during which thermodynamic quasi-equilibrium may be reached.

Despite these promising properties, all of the previous studies to create a dense gas of excitons in Cu$_2$O either in a bulk crystal or in a potential trap did not demonstrate conclusive evidence for an excitonic BEC [4, 10–14]. The main reason for this failure turned out to be the existence of a very efficient exciton–exciton annihilation process that sets in at high exciton densities and whose rate scales with the square of the exciton density $n$

$$\frac{dn}{dt} = -an^2. \quad (2)$$

The two-body decay constant $a$ was determined in the first report [15] to be of the order of $1 \times 10^{-20} \text{cm}^3\text{ns}^{-1}$ and was interpreted to be due to Auger-like scattering, whereby two excitons collide, the energy of one of the excitons being transferred by Coulomb interaction to the other and thus effectively ionizing this electron–hole pair into high conduction and valence band states. Therefore, this process does not only limit the exciton densities but also leads to a strong heating of the excitons, thereby preventing the cooling of the exciton gas down to the lattice temperature. In the meantime, there have been a number of both experimental and theoretical reports on this exciton annihilation process, but at present no conclusive understanding has been obtained. Instead, it turned out that the different reports quite disagree on both the magnitude and temperature dependence of this process. The values reported range from almost temperature-independent $7 \times 10^{-17} \text{cm}^3\text{ns}^{-1}$ [16, 17] to an inverse temperature-dependent rate $1.8 \times 10^{-16} \text{cm}^3\text{ns}^{-1}$ at $T = 77$ K [18]. In contrast, recent studies report an almost temperature-independent rate of $4 \times 10^{-16} \text{cm}^3\text{ns}^{-1}$ in bulk crystals [19]. Furthermore, the experimental results differ by at least four orders of magnitude from the theoretical predictions [18, 20].

To solve this discrepancy, recently it was suggested that the two-body decay process is actually related to the formation of a biexciton state [21]. However, in view of the disagreeing values in samples of different origin, a relation to impurities, which would relax the momentum and parity conservation due to the band symmetry of the pure crystal, seems to be plausible [18].

By a simple calculation it can be shown that with such large values of $a$ it will be impossible to reach exciton densities above $10^{17} \text{cm}^{-3}$ without applying laser powers above 100 mW, a limit which is set by the otherwise excessive heating of the crystal [10]. Therefore, the recent claim to have observed the transition into a condensed state for paraexcitons trapped in an external potential [14] by a relaxation explosion in a condensate similar to spin-polarized atomic hydrogen [22, 23] due to this Auger process is highly questionable, at least in view of experiments which seem to indicate the complete absence of the Auger process for paraexcitons in weak strain-induced potential traps [24].

Another problem in previous investigations has been the determination of reliable absolute exciton numbers. Here two methods are reported: one requires us to measure the interexcitonic transitions $1S \rightarrow nP$ [25, 26]. However, this requires access to the exciton cloud by mid-infrared radiation, which in our experiments would lead to excessive heating of the mixing chamber in our cryostat by thermal radiation. The other method calculates the exciton numbers from experimentally measured absolute intensities of the excitonic luminescence processes and relies on the known radiative rates [17]. While this method works quite well for phonon-assisted luminescence lines, i.e. for orthoexcitons, the determination of paraexciton numbers by the zero-phonon (ZP) emission processes remained problematic, because it was found that the conversion factor between intensities and exciton numbers does not show the expected strong dependence.
on exciton temperature, but could be taken as constant [24]. In this paper, we show that this puzzle can be solved by a theory of excitonic luminescence which takes into account the lifetime broadening of the exciton states.

To shed light on this situation and to explore the possibility of BEC in this system, we have started investigating the dynamics of the yellow excitons in cuprous oxide under the following new aspects: firstly, reduction of the temperature of the surrounding bath to as low as possible, secondly, the use of the purest available crystals, and thirdly, confining the excitons in a potential trap. In our studies we tried to realize traps with a rather small depth, as in previous studies the two-body decay constant $a$ was found to increase strongly with trap depth [24]. From the analysis of our results, we have to conclude that under our experimental conditions the two-body decay constants for paraexcitons are indeed much smaller than previously reported, while those for orthoexcitons are of the same order as those obtained in earlier studies. Furthermore, it is possible to collect paraexciton numbers well above $10^{10}$ during the relaxation pathway of the excitons.

The paper is organized as follows. In section 2, we present the details of our experimental setup and discuss some experimental problems. In section 3, we present our theory of excitonic luminescence. The next section discusses the procedure for analysing the experimental results. In section 5, we present the experimental results, which will be analysed and discussed with respect to various excitonic relaxation and decay processes. In section 6 we briefly discuss the possibility of an excitonic Bose–Einstein condensation in our experiments. The paper closes with a short summary and an outlook to further experiments.

2. Experimental procedures

Most important issues have been the following experimental measures:

1. The use of natural cuprous oxide crystals originally found in Namibia. From these crystals millimetre-sized cubic-shaped specimens with well-defined facets were prepared (see [27] for details). The quality of these samples was checked according to a low defect density, leading to long paraexciton lifetimes up to $1 \mu$s. For such samples, previous high-resolution absorption measurements in a magnetic field revealed a paraexciton line width as narrow as 80 neV, demonstrating their extremely high quality [28].

2. Reduction of the temperature of the surrounding bath as much as possible. For that purpose we employed a specially designed $^3$He/$^4$He dilution cryostat (Oxford Instruments) with direct optical access through two quartz windows for exciting excitons and detecting their optical emission with high spatial resolution (see figure 1). For the base temperature with closed windows, which will set the ultimate limit of reducing the bath temperature, we found 25 mK. However, due to excessive heat transfer from the environment through the optical windows, in the present experiments we could achieve only a minimum temperature of 820 mK. Here modifications of the window configurations that should reduce this heat flow and thus allow us to undercut this temperature are possible and under way.

3. Confinement of the excitons in a potential trap. The confinement is necessary to prevent diffusion of the exciton gas out of the excitation spot and thus changing the dimensions of the exciton cloud in time. To create the exciton potential trap, we use the well-known Hertzian stress technique [10, 13, 29, 30], where a spherical stressor made of glass (radius 7.75 mm) is pressed with a force $F$ against a flat surface of the crystal along a direction we
Figure 1. The main features of the experimental setup. Excitons are created by a dye laser propagating along the $x$-direction normal to the strain axis. The laser was focused either directly into the trap or onto a spot close to the trap. In the latter case, the excitons diffuse towards the trap due to the gradient force initiated by the potential trap, as indicated in figure 2. The emission out of the trap is monitored spatially resolved along the $z$-direction, while integrating along a small stripe along the $y$- and along the $x$-direction.

denote as the $z$-direction (figure 1). As a result, a confining potential in which the energies of ortho- and paraexcitons are lowered compared to bulk is generated. The equipotential contours in the $y, z$-plane for paraexcitons are shown in figure 2. This potential $V_{\text{ex}}(x, y, z)$ can be calculated from the known strain parameters of the yellow exciton states (for a recent calculation with refined parameters, see [27]). However, comparing the low-energy border lines of the measured spatio-spectral intensities, which should closely follow the potential profile, with calculated potentials (see e.g. the full lines in figure 4), we found the calculated spatial profiles to be systematically narrower irrespective of the parameters used for the calculation. By checking the surfaces of the stressor lens and the specimen, we could exclude any mechanical deformation due to excessive stress. Therefore, we suspect that the finite size of the stressor, neglected in the usual strain calculation, is responsible for this effect. As an alternative, we have fitted the experimental spectra with a Morse-type potential along $z$, $V(z) = V_0(\exp(-2\beta(z - z_0)/z_0) - 2 \exp(-\beta(z - z_0)/z_0))$, with minimum $-V_0$ at $z = z_0$, which can also be approximated by a parabolic function $V(z) = \alpha_1(z - z_0)^2 - V_0$ with the curvature parameter $\alpha_1 = 2\beta^2V_0/z_0^2$, and a two-dimensional harmonic oscillator normal to $z$ in the $x, y$-plane, $V(x, y) = \alpha_\perp(x^2 + y^2)$ (see figure 3(B)). The typical parameters for the para- and orthoexciton traps in the experiments described in this paper are given in table 1. We also found that in order to obtain the measured potential by the Hertzian theory, we had to increase the nominal radius of the stressor lens by a factor of 1.5, which is similar to that reported in [27].
4. Employment of a pulsed laser source with an almost optimum pulse width of 50 ns to create the excitons and a sufficiently low repetition rate (1 kHz) in order to keep the heat load as small as possible. We used a pulsed Q-switched Nd:YLF laser (Quantronix) pumped dye laser system (Radiant Dyes, optical line width 1 GHz), with which the excitation photon energy could be tuned around the excitonic absorption edge. Using a pulsed source has the further advantage that one can obtain from time-resolved measurements a complete picture of the relaxation dynamics of the exciton system.

5. Resonant creation of the excitons with as low excess kinetic energy as possible directly inside the trap. As shown in figure 3(A), there exist several excitation pathways to load the trap with paraexcitons. The first one uses excitation of orthoexcitons via resonance with the quadrupole-allowed ZP transition (O-ZP, process I), the second one uses an indirect absorption process involving an odd parity optical phonon, the strongest one being the $\Gamma_3^-$-transition (process II). Due to the shear strain in the trap, a third process would be possible, the direct creation of paraexcitons via the ZP transition (process III) [31]. The optimum excitation process has to fulfil the following criteria: (i) it should have an
Figure 3. (A) Different processes for the excitation and detection of excitons in Cu$_2$O. $|xz\rangle$, $|yz\rangle$: orthoexciton states in the trap; I: direct, II: phonon-assisted excitation of orthoexcitons; transitions in green mark the direct decay process of paraexcitons and those in blue the phonon-assisted decay of orthoexcitons. The arrows indicate the fast relaxation of the orthoexcitons within several ps due to acoustic phonon scattering. (B) The details of the excitation process via the $\Gamma_3^-$-assisted orthoexciton absorption. The red line shows the trapping potential for a value of $z$ outside the trap minimum, the black line indicates the energy of the photons used for excitation minus the phonon energy and the blue curve gives the density of absorbed photons and of excitons for 60 nJ pulse energy.

absorption strength that allows us to absorb a substantial fraction of the incoming laser photons. This rules out process III because the paraexciton absorption at low stress is weak. (ii) The spatial region where the excitons are created should be as large as possible to reduce the initial exciton density and hence the loss by density-dependent decay. This clearly favours the phonon-assisted process II, because the O-ZP transition has a peak absorption coefficient of about (500–1000)$\text{cm}^{-1}$. (iii) The primarily excited orthoexcitons and the subsequently created paraexcitons should drift completely down into the centre of the trap, pulled by the gradient force of the potential. This would favour positioning the laser spatially and spectrally exactly at the trap centre; however, in this case the luminescence of orthoexcitons overlaps that of paraexcitons in the trap centre. So we always positioned the laser beam about 100 $\mu\text{m}$ away from the trap centre in the positive $z$-direction away from the stressor lens. In order to confine the primarily created orthoexcitons already in the trap and to avoid any excitation outside the trap, which may lead to loss of the excitons, we tuned the energy of the laser photons slightly ($\approx$0.5 meV) below the onset of the phonon sideband in the bulk (at 2048.56 meV). (iv) The primarily excited orthoexcitons should be detectable in the same spectral measurement as the para- and orthoexcitons inside the trap. Due to selection rules (see [32]) this requires the $k$-vector of the excitation beam along the direction of observation. As shown by the calculation of the density of orthoexcitons created by this process in figure 3(B), we were able to absorb more than half of the incoming laser photons and keep the maximum orthoexciton concentration well below $5 \times 10^{13} \text{cm}^{-3}$ for laser pulse energies of 60 nJ, thus avoiding any two-body decay of these excitons.
Table 1. Parameters of the trapping potentials and of the rate model giving the best fit to the data.

| Parameter                              | Value          |
|----------------------------------------|----------------|
| Paraexciton trap depth                 | 3.338 meV      |
| Paraexciton $\alpha_\parallel$         | $0.155 \mu$eV $\mu$m$^{-2}$ |
| Paraexciton $\alpha_\perp$             | $0.078 \mu$eV $\mu$m$^{-2}$ |
| Orthoexciton trap depth                | 7.505 meV      |
| Orthoexciton $\alpha_\parallel$        | $0.255 \mu$eV $\mu$m$^{-2}$ |
| Orthoexciton $\alpha_\perp$            | $0.099 \mu$eV $\mu$m$^{-2}$ |
| Lifetime paraexcitons                  | 650 ns         |
| Ortho–para-conversion rate             | $0.2 \text{ ns}^{-1}$a |
| Two-body decay rate $a_{pp}$            | $3.7 \times 10^{-18} \text{ cm}^3 \text{ ns}^{-1}$ |
| Two-body decay rate $a_{OO}$            | $4.9 \times 10^{-17} \text{ cm}^3 \text{ ns}^{-1}$ |
| Relaxation rate $\Gamma_{rel}$          | $6 \times 10^7 \text{ s}^{-1}$ |
| Kinetic energy of excited orthoexcitons| 4.0 meV        |

a Taken from [24].

In the experimental setup (figure 1), the emitted light was imaged onto the entrance slit of a high-resolution triple spectrograph (T64000, Jobin Yvon) usable in either subtractive or additive mode with a diffraction limited spatial resolution of the order of 10 $\mu$m. The astigmatism of the spectrograph was corrected by a cylindrical lens (focal length $F = -1000 \text{ mm}$ or $F = 1000 \text{ mm}$ for subtractive and additive dispersion, respectively) in front of the entrance slit [27]. For a spatially resolved spectrum $I(z, \omega)$ along the direction of the applied strain, the luminescence from a small stripe centred in the trap, with width $2\Delta y$, was integrated along the $x$-direction perpendicular to $z$ (see figure 1). Detection was done by an intensified charge-coupled device (ICCD) camera (Andor iStar), which could be gated with a minimum temporal resolution of 5 ns for pulsed measurements. We measured both the spectral and spatial dependences of the excitonic luminescence, from which we could deduce the absolute luminescence intensities of the exciton states, the effective temperature and the spatial extent of the exciton cloud as a function of time. From these data we could determine the number of para- and orthoexcitons as a function of time in their potential traps with high accuracy. This allows us to test the relaxation models proposed very effectively and thus deduce accurate values for the various relaxation rates.

A typical spatially and spectrally resolved photoluminescence measurement obtained under the optimized conditions is shown in figure 4. In the upper part, the gate of the ICCD is set to a width of 200 ns centred at the laser pulse, whereas the lower part shows the time-integrated intensity over a gate width of 5 $\mu$s after the laser pulse has gone.

The spot designated $\text{Exc}$ represents the primarily excited orthoexcitons, observed via the $\Gamma_3^{-}$ phonon emission. These excitons relax into the trap, forming the band designated $\text{O}$. The first steps of the relaxation are visible in the spectrum as a sideband above $E_0$ shifted to smaller energies directly below the excitation spot. After this first step the orthoexcitons drift into the trap minimum, with their luminescence directly reflecting the spatial variation of the trap potential. The third band $\text{P}$ is stemming from the paraexcitons with their quite different trapping potential. These paraexcitons are formed by a rapid conversion process from the orthoexcitons
Figure 4. Spatially and spectrally resolved photoluminescence of ortho- and paraexcitons after excitation with a 45 ns laser pulse resonant into the phonon sideband of the orthoexciton in the trap. The spatial coordinate corresponds to a stripe along the direction of applied stress in the middle of the trap (z-axis). Exc: primarily excited orthoexcitons; O: orthoexcitons in the trap; P: paraexcitons in the trap. Note that the paraexcitons are observed via the ZP transition, while the orthoexcitons are observed via the $\Gamma_1^{-3}$ phonon sideband so that they lie spectrally below the paraexcitons. Upper panel: snapshot with 200 ns gate width during excitation pulse. Lower panel: snapshot starting 200 ns after the laser pulse with a gate width of 5 $\mu$s. The energy scale is given with respect to the paraexciton energy at $k = 0$ in the bulk $E_{0\text{Para}} = 2020.718$ meV, so that the paraexciton trap depth can be read off directly. For the orthoexcitons the difference between phonon energy $\hbar\Omega_1(\Gamma_1^{-3}) = 13.7$ meV and exchange splitting $\Delta = 12.1$ meV of 1.6 meV has to be added to obtain the trap depth. The full lines give the trapping potential for the ortho- and paraexcitons, which follow smoothly the bottom of the corresponding exciton luminescence. In the upper panel the arrows marked by $z_0$, $z_L$ and $E_0$ indicate the regions of integration to obtain the z-integrated spectra $I_{1-4}$ (see text). These regions are also marked at the right-hand side of the spectrum in the upper panel.

via a phonon-assisted spin-flipping process, which has been investigated quite intensively by different groups in the last decade [24, 33]. The rate of this conversion is 0.3 ns$^{-1}$ for bulk excitons and somewhat lower for excitons in the trap due to the smaller difference in energies. These paraexcitons still have a substantial amount of kinetic energy (about 8 meV) which they have to lose by emission of LA phonons. These relaxation processes might already lead to some
heating of the sample (if one excites $10^{10}$ excitons, the total amount of energy loss would be about $3 \times 10^{-10}$ J). At $T = 0.5$ K, this would lead to a temperature rise of the excited volume (diameter 100 $\mu$m) of about 2 K (with specific heats taken from [34]).

At later times, the excitons have thermalized in their trap and the paraexcitons show a characteristic high-intensity peak at the low-energy side, while the number of orthoexcitons is already reduced due to their shorter lifetime. From these data, the trap parameters such as depth $V_0$ and $\alpha_{\perp, \parallel}$ can be deduced. The results obtained for the present experiment are shown by the white lines and show excellent agreement.

3. Spectroscopy and thermodynamics of excitons in a potential trap: the ideal Bose gas

In the following, we describe the excitons as an ideal Bose gas with temperature $T$ and assume the validity of the semiclassical approximation [35]. Furthermore, we consider only the case of a noncondensed system with $T$ above the critical temperature $T_c$ for BEC $k_B T_c = \hbar \Omega_0 (N/\xi(3))^{1/3}$, where $\Omega_0 = \sqrt{2(\alpha_{\parallel} \alpha_{\perp}^2)^{1/3}/M_X}$ with $M_X$ being the exciton mass. For the thermodynamics we can neglect the exciton–exciton interaction since it would provide only a small correction to the external potential [35]. Then the distribution function is given by

$$f_{eq}(\vec{k}, \vec{r}) = \frac{1}{\exp\left(\frac{\hbar^2 k^2/2 M_X + V_{ex}(\vec{r}) - \mu}{k_B T}\right) - 1},$$

(3)

where $\mu$ is the chemical potential. For the external potential we use a functional form

$$V_{ex}(\vec{r}) = \alpha_{\parallel} (z - z_0)^2 + \alpha_{\perp} (x^2 + y^2),$$

(4)

whereby all energies are referred to the trap bottom at $-V_0$.

With the quantum degeneracy concentration

$$n_Q(T) = \frac{1}{4\pi^2} \left(\frac{2 M_X k_B T}{\hbar^2}\right)^{3/2},$$

(5)

the average trap volume

$$V_Q(T) = \pi \left(\frac{(k_B T)^3}{\alpha_{\parallel} \alpha_{\perp}^2}\right)^{1/2},$$

(6)

the Bose integrals

$$\eta(a, n) = \int_0^\infty dx \frac{x^n}{e^{x+a} - 1},$$

(7)

$$Q(a, n) = \int_0^\infty dx x^n \eta(x + a, 1/2),$$

(8)

$$H(a, n) = \int_0^\infty dx \frac{x^n}{e^{x+a} - 1} \eta(x + a, 1/2),$$

(9)

the exciton density is given by

$$n(\vec{r}) = n_Q(T) \eta\left(\frac{-\mu - V_{ex}(\vec{r})}{k_B T}, 1/2\right),$$

(10)

and the total number of particles by

$$N = n_Q(T) V_Q(T) Q\left(\frac{-\mu}{k_B T}, 1/2\right).$$

(11)
3.1. Direct (zero-phonon) emission process

Inside the traps the paraexcitons recombine by a direct emission of a photon due to mixing to the green exciton states [36]. The spectrally resolved luminescence intensity with energy $h\omega$ (referring to the energy of the trap minimum) in the direction of $\vec{k}_0$ (solid angle $d\Omega$) stemming from a volume $dV$ for the ZP process is given by [35]

$$dI_{\text{ZP}}(h\omega, \vec{r}) = \frac{S_0}{(2\pi)^3} A_{\text{id}}(\vec{k}_0, \hbar \omega_{\text{loc}}(\vec{r}) - \mu_{\text{loc}}(\vec{r})) n_B(\hbar \omega_{\text{loc}}(\vec{r}) - \mu_{\text{loc}}(\vec{r})) dV d\Omega$$

(12)

assuming that only excitons with wavevector $k_0 = n_B 2\pi / \lambda_0$, where $n_B$ is the Bose function, interact with strength $S_0$ with light. $\hbar \omega_{\text{loc}}(\vec{r}) = \hbar \omega - V_{\text{ex}}(\vec{r})$ is the local photon energy and $\mu_{\text{loc}} = \mu - V_{\text{ex}}(\vec{r})$ is the local chemical potential. $A_{\text{id}}(\vec{k}_0, \hbar \omega) = 2\pi \delta(\hbar \omega - E(\vec{k}_0))$ with $E(\vec{k}) = \hbar^2 k^2/2M + V_{\text{ex}}(\vec{r}) - \mu$ is the spectral function of the ideal Bose gas [35, 37]. In order to obtain a simple analytical expression, we will neglect the spatial variation of the radiative rate due to the variation of the local strain. For small strain and low temperatures this still gives an accurate description, but any spatial dependence can be easily incorporated into a numerical calculation.

However, equation (12) does not take into account the damping of the emitting states and does not fulfill the fundamental relation between absorption and emission [38]. One can correct this error by taking into account the uncertainty of the wavevector due to the lifetime broadening of the exciton states [39, 40]. If $h\gamma$ is the lifetime broadening of the exciton state at $\epsilon_0 = \hbar^2 k_0^2/2M$ with $M$ being the paraexciton mass, then the uncertainty in the wavevector is simply given by $\Delta k = \sqrt{M/2E_0} \cdot \gamma$, and the density of the states contributing to the luminescence is just

$$D'_{\text{ZP}} = \frac{1}{(2\pi)^3} \int d\Omega k_0^2 \Delta k = \frac{k_0 M}{2\hbar^\gamma \cdot \gamma} = D_0 \gamma$$

(13)

with which (12) has to be multiplied to obtain the proper intensity. For the exciton damping $\gamma$ two contributions are important [28]: scattering by acoustical phonons, which leads to a contribution $\gamma_0$ that depends only on the temperature of the surrounding phonon bath, and exciton–exciton scattering. The latter contribution $\gamma_{XX}$ can be assumed to depend on the local exciton density $n(\vec{r})$, the mean exciton velocity $\bar{v} = \sqrt{18k_B T/(\pi M)}$ and the scattering cross section $\sigma_{XX} = 8\pi a_0^2$ as [28]

$$\gamma_{XX} = \sigma_{XX} n(\vec{r}) \bar{v}(T).$$

(14)

Integrating over the solid angle and over the $x$-direction, we then obtain as spatio-spectral intensity

$$I_{\text{ZP}}(h\omega, y, z) = \frac{S_0 \cdot D_0}{4\pi^2} n_B(h\omega - \mu) \Theta(\chi_0) \left[ \gamma_0 + \sigma_{XX} \bar{v} n_Q(T) \eta \left( \frac{\mu - h\omega + \epsilon_0}{k_B T}, 1/2 \right) \right]$$

(15)

with $\Theta$ being the Heaviside step function, $\chi_0 = \sqrt{(\hbar \omega - \epsilon_0 - a_0 z^2 - a_{3\perp} y^2)/\sqrt{a}}$. We see that the damping enters this expression via a spatially independent, but spectrally dependent, term

$$D_{\text{ZP}}(h\omega, T, \mu) = D_0 \left[ \gamma_0 + \sigma_{XX} \bar{v} n_Q(T) \eta \left( \frac{\hbar \omega - \epsilon_0 - \mu}{k_B T}, 1/2 \right) \right],$$

(16)

which has to be multiplied to all the following expressions.

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From this result, we can obtain the spatially resolved spectrum \( I_{ZP}(\hbar \omega, z) \) by integrating over a slit with width \( 2 \Delta y \) in the \( y \)-direction as

\[
I_{ZP}(\hbar \omega, z) = \frac{S_0 \cdot D_{ZP}(\hbar \omega, T, \mu)}{4\pi^2} n_B(\hbar \omega - \mu) \frac{2}{\alpha_\perp} \times \arcsin \left[ \min \left( 1, \sqrt{\frac{\alpha_\perp \Delta y^2}{\hbar \omega - \varepsilon_0 - \alpha_\parallel z^2}} \right) \right] \cdot \Theta(\hbar \omega - \varepsilon_0 - \alpha_\parallel z^2),
\]

which will be used to fit the spatio-spectral intensities.

The spectral profile \( I_{ZP}(\hbar \omega) \) can be obtained by integrating (17) over \( z \) to

\[
I_{ZP}(\hbar \omega) = \frac{S_0 \cdot D_{ZP}(\hbar \omega, T, \mu)}{4\pi \sqrt{\alpha_\perp \alpha_\parallel}} n_B(\hbar \omega - \mu) \min \left( \Delta y, \sqrt{\frac{\hbar \omega - \varepsilon_0}{\alpha_\perp}} \right).
\]

The total integrated intensity is given by

\[
I_{ZP} = \frac{S_0 \cdot D_{ZP}(\hbar \omega, T, \mu)}{4\pi \sqrt{\alpha_\perp \alpha_\parallel}} \int_0^\infty n_B(u - \mu + \varepsilon_0) \cdot D_{ZP}(u, T, \mu) \sqrt{\min(\alpha_\perp \Delta y^2, u)} du.
\]

In the limit of large slitwidth \( \Delta y \to \infty \), we obtain as total integrated intensity

\[
I_{ZP,\text{tot}} = \frac{S_0 \cdot D_0}{4\pi^2} V_Q(T) \left[ \gamma_0 \eta \left( \frac{-\mu - \varepsilon_0}{k_B T}, 1/2 \right) + \sigma_{XX} \bar{n}_Q(T) H \left( \frac{\mu - \varepsilon_0}{k_B T}, 1/2 \right) \right],
\]

which is related to the number of excitons in the trap via the chemical potential by (11).

By integrating (15) over \( \hbar \omega \) we obtain simple expressions only for low density, i.e. if \( \gamma \simeq \gamma_0 \). The spatially resolved spectrally integrated intensity \( I_{ZP}(x, z) \) is given by

\[
I_{ZP}(x, z) = \frac{S_0 \cdot D_0 \gamma_0}{4\pi^2} \frac{k_B T}{\alpha_\perp} \eta \left( \frac{-\mu - \varepsilon_0 - \alpha_\parallel z^2 - \alpha_\perp x^2}{k_B T}, -1/2 \right)
\]

and the \( z \)-profile by integrating (21) over the slit

\[
I_{ZP}(z) = \frac{S_0 \cdot D_0 \gamma_0}{2\pi^2} \frac{k_B T}{\alpha_\perp} q \left( \frac{\alpha_\perp \Delta y^2}{k_B T}, -\mu + \varepsilon_0 + \alpha_\parallel z^2 \right)
\]

with the abbreviation

\[
q(x, a) = \int_0^x \eta(u + a, -1/2) du.
\]

From both relations (19) and (20) we see that the integrated intensity of the ZP process depends not only on temperature, but also on the exciton density via exciton–exciton collisions increasing
the exciton damping $\gamma$. For pulsed excitation, we expect both the number of excitons and the temperature to decrease, thus allowing no simple relation between the ZP intensity and the number of excitons. For practical applications this has two important consequences:

1. The high-energy tail of the luminescence is no longer given by a Bose distribution, but one has to take into account an additional temperature and density-dependent correction. This can only be neglected in the case of a very dilute Bose gas when $\gamma \approx \gamma_0$ is constant, i.e. at very low particle numbers in the trap or at high enough exciton energies. We therefore avoided in the analysis, whenever possible, determining absolute exciton numbers from the integrated intensity of the paraexciton ZP transition alone.

2. The spatial extension of the exciton cloud is no longer given by simple Gaussian functions (see (31)), but may differ considerably.

3.2. Phonon-assisted transitions

In a similar way the lineshape of the phonon-assisted transition can be obtained by taking into account that now all exciton states contribute to the luminescence intensity. Therefore,

$$I_{PH}(\hbar \omega, \vec{r}) = S_{PH}D_{loc}n_B(\hbar \omega - \hbar \Omega - \mu)$$

with $\Omega$ being the phonon frequency and $D_{loc}$ the local density of states given by

$$D_{loc}(\hbar \omega, \vec{r}) = \frac{1}{4\pi^2} \left( \frac{2M_X}{\hbar^2} \right)^{3/2} E_{loc} \Theta(E_{loc})$$

with $E_{loc} = \hbar \omega' - \alpha_\parallel z^2 - \alpha_\perp (x^2 + y^2)$ and $\hbar \omega' = \hbar \omega - \hbar \Omega$. Integrating over the $x$-direction results in

$$I_{PH}(\hbar \omega, y, z) = \frac{S_{PH}}{8\pi} \left( \frac{2M_X}{\hbar^2} \right)^{3/2} \frac{\hbar \omega' - \alpha_\parallel z^2 - \alpha_\perp y^2}{\sqrt{\alpha_\perp}} n_B(\hbar \omega' - \mu).$$

Integrating this quantity over the slit results in

$$I_{PH}(\hbar \omega, z) = \frac{S_{PH}}{4\pi} \left( \frac{2M_X}{\hbar^2} \right)^{3/2} n_B(\hbar \omega' - \mu) \cdot (\hbar \omega' - \alpha_\parallel z^2)$$

$$\times \min \left( \frac{2/3}{\hbar \omega' - \alpha_\parallel z^2}, \left( \frac{\alpha_\perp \Delta y^2}{\hbar \omega' - \alpha_\parallel z^2} \right)^{1/2}, \left( \frac{\alpha_\perp \Delta y^2}{\hbar \omega' - \alpha_\parallel z^2} \right)^{3/2} / 3 \right).$$

Finally, the integrated spectrum is obtained as

$$I_{PH}(\hbar \omega) = \frac{4S_{PH}}{\pi} \left( \frac{2M_X}{\hbar^2} \right)^{3/2} \frac{\hbar \omega'}{15 \sqrt{\alpha_\parallel \alpha_\perp}} q \left( \min \left( 1, \frac{\alpha_\perp \Delta y^2}{\hbar \omega'} \right) \right)$$
with \( q(x) \) given by

\[
q(x) = \int_0^x (1 - x'^2)^{3/2} \, dx'.
\]  

Integrating over \( h\omega' \), we obtain the total intensity of the phonon sideband as

\[
I_{PH} = \frac{4S_{PH}}{15\pi \sqrt{\alpha_\perp^2 \alpha_\parallel}} \int_0^\infty \left( \frac{2M_X}{\hbar^2} \right)^{3/2} n_B (u - \mu) \cdot u^2 q \left[ \min \left( 1, \sqrt{\frac{\alpha_\perp \Delta y^2}{u}} \right) \right] \, du.
\]  

In contrast to the ZP transition, the intensity of the phonon sideband depends not on the homogeneous broadening of the exciton states, but due to the temperature dependence of the size of the exciton cloud on the slit width alone. For small slits it goes as \( T^{-1/2} \), while for wide slits, when the complete trap is imaged onto the slit, it stays constant. As an important consequence, the ratio between the ZP luminescence band and the phonon-assisted sideband depends not only on the ratio of the radiative transition probabilities but also strongly on temperature and exciton density. Therefore, the determination of exciton numbers via a phonon-assisted band is much more reliable and should be preferred.

4. Procedure of analysis of the experimental results

As our goal is a quantitative analysis of the experiments, we have to consider carefully the procedure to obtain the absolute numbers of excitons. This is done by applying the quantitative emission theory outlined in the foregoing section for each time step. For this purpose, we have to determine the potential profiles, the spatial extension and the temperature of the exciton clouds.

4.1. Potential profiles, spatial size and exciton temperature

According to the previous section, the determination of exciton numbers requires several quantities, such as the radiative coupling strength, the exciton temperature and the size of the exciton cloud.

The spatial extensions of the excitons depend on the potential curvatures and on temperature. According to (15), in the case of a chemical potential far away from the trap bottom, i.e. low enough exciton numbers, one obtains simple Gaussian functions for the intensity profiles

\[
I(y, z) = I_0 \exp \left( -\frac{y^2}{\sigma_\perp^2} \right) \exp \left( -\frac{z^2}{\sigma_\parallel^2} \right)
\]  

with the size parameters \( \sigma_\parallel, \sigma_\perp \) differing in the \( z \)- and \( y \)-directions reflecting the different potential shapes. If the exciton gas is in thermal equilibrium with temperature \( T \), the size parameters are given by

\[
\sigma_{\perp, \parallel} = \sqrt{\frac{k_B T}{\alpha_{\perp, \parallel}}}.
\]  

From spatio-spectral profiles like those in figure 4, we can directly determine the potential shape in the \( z \)-direction and from this derive the curvature parameter directly. These values are
given in table 1. We can check the consistency of our analysis by comparing the experimentally determined size parameters with those from (32). The results in figure 8(B) show satisfactory agreement. To obtain the curvature parameters in the xy-plane, one either can turn the image on the slit by 90° or, much simpler, take advantage of the subtractive mounting of the entrance stage of the triple monochromator. By setting the third spectrographic stage to zero wavelength, we can monitor the spectrally integrated distribution of the luminescence in the yz-plane, with the spectral window set by the intermediate slit of the subtractive stage. This distribution corresponds directly to the spatially resolved intensity given by (15) and (26) for the ZP and phonon process, respectively, from which we can determine $\alpha_\perp$ by fitting the appropriate Gaussian (31). The values obtained in this way correspond very well to those obtained by calculating the stress-induced potential shape with modified lens radii.

The spatial cloud of the primarily excited excitons can be assumed to follow the spatial profile of the excitation laser, which is a Gaussian beam in the x, z-plane with variance $\sigma_\perp$ and can be determined by analysing the excitation spot.

The effective temperature of the exciton cloud can be obtained straightforwardly by fitting the high-energy tail of the spatially integrated intensity (18) of the ZP line, since the curvature parameter $\alpha_\perp$ only influences the low-energy lineshape.

4.2. Determination of the number of excitons in the trap

In order to derive quantitative relaxation rates, one has to obtain from the luminescence measurements the absolute numbers of the various exciton species in the trap. We apply here the method of measuring absolute luminescence intensities which has been demonstrated to obtain numbers within reasonable error bars [17]. It relies on the fact that the total rate of photons emitted in all spatial directions of an excitonic line is simply given by the number of excitons times the radiative rate of this process.

Therefore, the first step consists in the estimation of the total rate of emitted photons from the experimentally detected number of counts of the CCD detector integrated over the luminescence line under consideration. If we have inside a volume $dV$ at time $t$ and position $\vec{r}$ a number of excitons radiating $dN_{\text{phot}}$ photons during the time interval $dt$ in all directions of space, the total number of counts of the CCD detector due to photons emitted by an exciton cloud which is imaged onto the entrance slit of the spectrograph (slitwidth $2\Delta y$) with a magnification $M_\parallel$, $M_\perp$ parallel and perpendicular to the slit direction and reaching the detector is given by

$$I_{\text{det}}(z, E)(t) = R T_{\text{int}} \eta \int_0^{t+T_G} dt \int_{-\Delta y}^{\Delta y} dy \int_{-d/2}^{d/2} dx dN_{\text{phot}} \left( x, \frac{y}{M_\parallel}, \frac{z}{M_\perp}, t \right),$$

where we neglected the spatial and spectral blurring due to the finite resolution of the setup. Here $d$ is the sample thickness, $R$ the repetition rate of the excitation laser, $T_G$ the period in which the gate of the detector is open, $T_{\text{int}}$ the total integration time and $\eta$ the overall detection efficiency comprising the efficiency of the optical setup $\eta_{\text{det}}$ (losses in optical elements, spectrometer and the quantum efficiency of the ICCD detector), the finite solid angle for detection and the reflection losses of the cryostat windows and the sample.

Therefore $\eta$ is given by

$$\eta = (1 - R_{\text{Cu2O}})(1 - R_Q) 10^{-10} \frac{(\sin \alpha_0)^2}{4\pi n_0^2} \eta_{\text{det}}.$$

New Journal of Physics 14 (2012) 023054 (http://www.njp.org/)
Here $n_0 = 2.95$ is the index of refraction of $\text{Cu}_2\text{O}$ at 615 nm, $R_{\text{Cu}_2\text{O}} = (1 - n_0)^2 / (1 + n_0)^2$ and $R_Q = (1 - n_Q)^2 / (1 + n_Q)^2$ ($n_Q = 1.45$) are the Fresnel reflection losses of sample and quartz windows and $\theta_0 = 0.089$ is half the detection angle of the entrance lens (Schneider Apo-Symmar 5,6/210). Finally, the number of emitted photons per detector count $\eta_{\text{det}}$ was measured to be $\eta_{\text{det}} = (3.2 \pm 0.1)$. The magnification was determined from the image of a Ni grating at the position of the sample as in [27] to be $M_\parallel = 4.17$ and $M_\perp = 5.10$. By dividing the measured counts from the detector by $\eta$ and by the effective measuring time $T_{\text{meas}} = R \cdot T_G \cdot T_{\text{int}}$, we obtain the total rate of emitted photons averaged over the gate time, the quantity that is displayed in figure 8 and that is used to derive the number of excitons in the trap according to the procedure given above. It should be noted, however, that some of the equations derived include already the fact that in order to obtain a high spectral resolution, the entrance slit of the spectrograph had to be closed down to some $10 \mu m$, so that only a fraction of the exciton cloud is imaged. We therefore either used these results directly to determine the total number of excitons or used the measured spatial profiles to correct directly for the vignetting by the slit using Gaussian distributions (31) and used the relations (20) and (30) to determine the number of excitons. The results of both procedures agreed in most cases within 10%.

4.3. Radiative rates

One central quantity in the determination of exciton numbers is the radiative transition rate of the corresponding transition. Compared to the results of [41], we have to consider four points.

1. We detect the orthoexcitons via the $\Gamma_3^-$ phonon transition. While for the situation in the bulk, the detailed balance arguments in [17] are derived for unpolarized absorption and emission, we have to take into account that the orthoexcitons radiate from inside the potential trap in the [110] direction. The selection rules [32] then predict that only the $|xz\rangle$ and $|yz\rangle$ states radiate with a polarization in the $x$-direction, but with a probability that is $3/2$ of the average in the bulk, while the $|xy\rangle$ state would radiate only with polarization along the $z$-direction, but is shifted towards higher energies and not populated at all. Therefore, for the orthoexciton we set the radiative rate to be

$$\Gamma_{\text{rad}}(\text{ortho}, \Gamma_3^-) = (10.5 \pm 2.1) \times 10^4 \text{s}^{-1}. \quad (36)$$

2. The radiative rate for the $\Gamma_5^-$ phonon-assisted paraexciton transition in the bulk is given by [16]

$$\Gamma_{\text{rad}}(\text{para}, \Gamma_5^-) = (1.5 \pm 0.2) \times 10^2 \text{s}^{-1}, \quad (37)$$

which is given here only for reasons of completeness as we did not use the phonon-assisted paraexciton transition in our analysis.

3. As the radiative rate is defined as the ratio between the total number of emitted photons per second in the phonon sideband and the number $N$ of excitons:

$$\dot{N}_{\text{phot}} = \Gamma_{\text{rad}} N, \quad (38)$$

the following relation can be easily obtained:

$$\Gamma_{\text{rad}} = \lim_{\Delta y \to \infty} \frac{I_{\text{PH}}}{N} \quad (39)$$
from which $S_{PH}$ can be obtained using (30). For the paraexciton, where only the $\Gamma_5^-$ phonon is allowed, we obtain for the coupling strength $S_{PH} = (94 \pm 10) \, \mu eV^{3/2} \, s^{-1}$, while for the orthoexciton decay via the $\Gamma_3^-$ process $S_{PH} = (4.4 \pm 0.9) \times 10^3 \, \mu eV^{3/2} \, s^{-1}$.

4. The detection process for paraexcitons via the ZP transition has the advantage that it is not obscured by overlying decay processes of the trapped orthoexcitons. As shown in the previous section, the intensity of this process is determined both by the coupling strength $S_0$ and by the excitonic damping. This quantity has been extensively analysed by measurements of the paraexcitonic absorption as a function of temperature and exciton density [28, 42]. For a bath temperature $T = 0.82$ K, we can read off $\gamma_0 = 0.025 \, \mu eV$, while for the scattering length we use $a_S = 2.1 a_B = 1.5$ nm from a quantum Monte Carlo calculation [43]. The coupling constant $S_0$ depends on the square of the shear strain [36]. For small stress and low temperatures where the size of the exciton cloud is quite small, the variation is less than 10% and therefore will be neglected. We should note that, as we estimate $\Gamma_{\text{rad}}$ from above, our estimates for the exciton numbers will be systematically too small. In principle, it should be possible to determine the ZP coupling constant $S_0$ for the strain situation by comparing the total intensity of the phonon-assisted $\Gamma_5^-$ process with that of the ZP process. However, because of an accidental degeneracy, this band overlaps with the orthoexciton $\Gamma_4^-$ transitions and, due to its weakness, it could not be resolved separately. In the analysis, we therefore did not use the absolute intensity of the paraexciton transitions as the input parameter, but used it only as an a posteriori check of the consistency of the data. Indeed, by assuming a constant value of $S_0$ for all experiments in the same strain conditions, we are able to fit all spectra without any other assumptions, therefore allowing us to derive a consistent set of exciton numbers.

5. Experimental results

5.1. Power dependence and time-resolved spectroscopy

Full insight into the exciton relaxation dynamics can be obtained from series of time-gated measurements of the spatio-spectral luminescence with varying laser excitation power and covering the whole decay. However, to obtain sufficient signal-to-noise ratio, the required measurement times are too long to guarantee the stability and reproducibility of the setup. We therefore measured the power dependence at the two gate settings shown in figure 4 over two orders of magnitude and the time series only at selected power levels. In analysing the whole bunch of data, it turned out that the most important information is contained already in two quantities, the spatial profiles, integrated over the spectral coordinate for both ortho- and paraexcitons up to 2 meV above their trap minimum, and the spectra, obtained by integrating over the spatial dimension of the traps.

In order to obtain reproducible information from the spectra, we decomposed the measured spatio-spectral profiles into three uniquely defined components representing the primarily excited excitons, the orthoexcitons and the paraexcitons in the trap. This is possible by noting that the paraexciton potential crosses the excitation spot (centre at $z_L$) at some definite $z_0$ (see figure 4). Then for $z > z_0$, we can exclude any contribution from paraexcitons to the luminescence signal. Furthermore, in the spectral range below $E_0$ (see figure 4), no primarily excited orthoexcitons can be present. We therefore define four $z$-integrated spectra $I_i, i = 1 \cdots 4$, plotted in figure 5(A) in the following way: $I_1$ is the total integrated intensity, $I_2$ is the intensity integrated over $z > z_0$, $I_3$ is integrated over the excitation spot from
Figure 5. Decomposition procedure to obtain spectral lineshapes for the different exciton species. The excitation pulse energy was 15 nJ and the temperature of the liquid He bath was 0.82 K. (A) Exc, O, P denote the primarily excited orthoexcitons, the orthoexcitons in the trap and the paraexcitons in the trap. \( I_1 \cdots I_4 \) are the spectra obtained by integrating the spatially resolved spectrum over the \( z \)-ranges as discussed in the text. (B) Result of the decomposition into the spectra of total and relaxed para- and orthoexcitons in the trap \((I_1 - c_{12} I_2, I_4 - c_{42} I_2)\) and the spectrum of the primarily excited orthoexcitons \((c_{13} I_3 - c_5 I_{\text{hot}})\).

\[ z = 2z_L - z_0 \] to \( z = z_0 \), and finally \( I_4 \) is integrated from \( z = 0 \) to \( z = 2z_L - z_0 \) and represents the relaxed excitons in the trap (see figure 5(A)). Then we determine four constants \( c_{12}, c_{13}, c_{42}, c_5 \) by equating \( I_1 = c_{12} I_2, I_1 = c_{13} I_3 \) and \( I_4 = c_{42} I_2 \) in the spectral region just below the paraexciton line. Then the total number of para- and orthoexcitons is given by \( I_1 - c_{12} I_2 \), the fraction of relaxed paraexcitons and orthoexcitons is given by \( I_4 - c_{42} I_2 \), and the difference of these spectra \( I_{\text{hot}} \) represents the hot excitons in the corresponding trap. On the other hand, the primarily excited orthoexcitons are given by \( c_{13} I_3 - c_5 I_{\text{hot}} \), whereby \( c_5 \) is determined by the condition that at \( E < E_0 \) no primarily excited orthoexcitons exist. In order to keep the total number of counts conserved, we have to add the contribution \((c_5 - 1) I_{\text{hot}}\) to the excitons in the trap. In this way, we always obtain a unique decomposition of the measured spectra into the different exciton species (see figure 5(B)). The total integrated number of counts for the various exciton lines is finally obtained by summing the corresponding spectra over the appropriate energy range, e.g. for the orthoexcitons in the trap from \( E = -9.5 \) meV to \( E = -4.5 \) meV.
Figure 6. Spectral lineshapes of time-resolved luminescence of paraexcitons for different delay times, $t$, with respect to the laser pulse as indicated; the gate width is 200 ns. The red data points are given on the logarithmic scale on the left-hand side; the green data points refer to the linear scale on the right-hand side. The insets show the corresponding spatial profile along the stress direction, $z$. The spectral lineshapes are fitted by (18) with temperature $T_{\text{eff}}$ and exciton number $N_P$ as explained in the text. The excitation pulse energy was 15 nJ and the temperature of the liquid He bath was 0.82 K.

In figure 6, we show three spatial profiles and spectral lineshapes of the paraexciton $ZP$ transition obtained from a time-dependent series at rather low excitation power, which is typical for all of the results. At small delay times the exciton distributions are broad and the spectrum
Figure 7. Spectral lineshapes of measurements as in figure 4 for different laser excitation powers $P_L$ and gate width $\Delta t$ as indicated. Again, the red data points are given on the logarithmic scale on the left-hand side; the green data points refer to the linear scale on the right-hand side. The spectral lineshapes are fitted by (18) with temperature $T_{\text{eff}}$ and exciton number $N_p$ as explained in the text. The spectral lineshapes have a pronounced high-energy tail indicating a rather high temperature. For later times, the spatial extension shrinks and the energy distribution becomes much narrower. Both effects indicate a substantial cooling of the excitons during the relaxation process, details of which will be analysed in the following section. To get quantitative results, we fitted the spatial profiles by a Gaussian lineshape (see the insets) and the spectra by the lineshape function (18). The estimated temperatures are indicated in each panel. The overall good agreement allows us to ascribe a well-defined effective temperature to the excitons. Here we should also remark that the values of the size parameters agree very well with those expected from (32) using the potential parameters from table 1 if we take into account the finite spatial resolution ($\sigma_{\text{res}} = 18 \mu\text{m}$). The excellent agreement also shows that the excitons are in quasi-thermal equilibrium, a mandatory prerequisite for the application of the Bose gas formalism.

In figure 7, we plot a series of spectra for excitation with different power. Here we took spectra for the first 200 ns during the excitation pulse and for a gate window of $5 \mu\text{s}$ after the pulse as in figure 4. These data were analysed in the same way to obtain the effective temperature and the size of the exciton cloud. We find that the temperature decreases from about 10 K down to about 2 K with increasing time, being systematically higher for higher excitation powers.

In figure 8, the results of the analysis for different delay times are shown. Panel (A) shows the total emitted photon rate for the three different exciton states (exc, ortho, para) as a function of the delay time. While the primarily excited orthoexcitons have vanished almost completely
Figure 8. The results of the analysis of a series of time-dependent measurements, $T_{\text{bath}} = 0.82$ K, $P_L = 15 \mu$W. (A) Total emitted photon rate for primarily created orthoexcitons (open squares), orthoexcitons in the trap (blue dots) and paraexcitons in the trap (red squares). (B) The blue dots and red squares indicate the orthoexciton and paraexciton temperatures (left scale). The blue circles and red open squares mark the orthoexciton and paraexciton size parameters (right scale), which is just the full-width at half-maximum of the spatial distribution.

after 200 ns, the paraexcitons show a rather long decay time of several hundreds of ns. The orthoexcitons decay much more slowly than expected from the short ortho–para conversion time of about 5 ns inside the trap, indicating already the existence of a two-particle decay process leading to regeneration of ortho- from the paraexcitons known from the literature [17, 24].

Panel (B) shows the temporal dependence of the experimentally determined effective exciton temperatures and size parameters. The temperatures can be fitted quite well by a cooling curve of the form

$$T(t) = (T_{\text{ini}} - T_{\text{fin}}) \exp(-t/\tau_c) + T_{\text{fin}}$$

(40)

with the final temperature equal to the bath temperature, an initial temperature of $T_{\text{ini}} = 12.5$ K and a cooling time of $\tau_c = 220$ ns as shown by the dotted lines in the figure. The cooling down of the exciton gas is reflected also in the size of the exciton cloud, which shrinks from 120 $\mu$m down to about 35 $\mu$m. As we saw in figure 7 the initial temperature depends weakly on the excitation power, but in all cases the excitons reach the same final temperature close to the bath temperature, i.e. the temperature of the mixing chamber of 0.82 K.

5.2. Rate equation analysis

Description of the temporal dependence of exciton numbers by rate equations is a well-established method and has been applied previously also to studies of yellow excitons (see e.g. [15, 19, 21, 24]). Due to the rather special situation in the case of trapped excitons, the rate equations simplify considerably, which makes the subsequent analysis much more reliable. As was already pointed out, we have to distinguish three different exciton species, the primarily

New Journal of Physics 14 (2012) 023054 (http://www.njp.org/)
excited orthoexcitons, the relaxed orthoexcitons in the trap and the paraexcitons in the trap. Excitons that are outside the trap are of very low concentration and can be neglected. Therefore, we consider the following processes:

1. Excitation of orthoexcitons with number $O_{\text{exc}}$ by a laser pulse with temporal profile $G(t)$, which can be well described by

$$G(t) = \frac{1}{1.772 T_p} \exp \left( -\frac{(t/T_p)^2}{2} \right), \quad (41)$$

where $T_p = 25$ ns is the pulse duration. The total number of absorbed photons is given by the parameter $N_{\text{abs}}$. Here we can safely assume that each photon absorbed leads to the creation of one exciton.

2. Conversion of these excited orthoexcitons into trapped excitons with a rate $\Gamma_{\text{rel}}$ by relaxing down the trap potential.

3. Conversion of orthoexcitons into paraexcitons by the density-independent acoustic phonon-assisted process [33]. This conversion is effective both in the trap and in the excitation spot. According to the recent investigations this rate depends weakly on the applied stress. In our case (stress 1.5 kbar) it can be taken to be $\Gamma_{\text{op}} = 0.2$ ns$^{-1}$. At the low temperatures of our experiment, we neglect the weak temperature dependence of this process which has been found recently [33]. We further include a possible density-dependent spin-flip process with rate $\Gamma_{\text{SF}}$ although this seems to be of minor importance at low temperatures [33].

4. Radiative and non-radiative decay of both ortho- and paraexcitons with total rates $\Gamma_O$, $\Gamma_P$, which determine their lifetimes.

5. Two-body decay of ortho- and paraexcitons. Usually this process is considered as an Auger-like recombination, where two excitons are destroyed by the transfer of one electron–hole pair into higher-lying valence and conduction band states. From these states the particles relax in energy and form again an exciton. Here we assume that all four exciton states are formed with equal probability. This means, however, that one of the orthoexciton states (designated normally as $|xy\rangle$) is not trapped and will drift into the bulk and subsequently gets lost. Therefore the regeneration factors that take into account this remaking of excitons have different values in the trap. Due to the varying exciton density in the trap, we measure for the two-body decay only a rate averaged over the exciton cloud. Assuming thermal distribution of the form of (10) we obtain the following relation to the volume rates:

$$A_i = \left( \frac{2\alpha_1\alpha_2^2}{\pi (k_B T)^3} \right)^{1/2} a_i \quad (42)$$

with $i=\text{PP, OO, OP}$ denoting the scattering species. Assuming the volume rates $a_i$ to be temperature independent [16, 17, 19], equation (42) results in a dependence of the form $A_i = A_0/(T/T_{\text{ref}})^{3/2}$, with an arbitrarily chosen $T_{\text{ref}} = 2.5$ K.

For the primary excited excitons, we can neglect the two-body decay, since the exciton density is much smaller because of the large excitation volume (see figure 3(B)).
Table 2. Meanings of the symbols in the rate model (43), (44) and (45).

| Symbol | Meaning |
|--------|---------|
| $O_{\text{exc}}$ | Primary excited orthoexcitons |
| $O_T$ | Trapped orthoexcitons |
| $P_T$ | Trapped paraexcitons |
| $N_0$ | Total absorbed photons |
| $G(t)$ | Temporal profile of the laser pulse |
| $\Gamma_{\text{rel}}$ | Relaxation rate into the trap |
| $\Gamma_{\text{OP}}$ | Ortho–para-conversion rate |
| $\Gamma_O$ | Orthoexciton decay rate |
| $\Gamma_P$ | Paraexciton decay rate |
| $\Gamma_{\text{SF}}$ | Spin–flip rate |
| $A_{\text{OO}}$ | Two-body decay rate for orthoexcitons |
| $A_{\text{PP}}$ | Two-body decay rate for paraexcitons |
| $A_{\text{OP}}$ | Two-body decay rate for ortho- and paraexcitons |

This gives the following set of rate equations:

\[
\frac{dO_{\text{exc}}}{dt} = N_0 \cdot G(t) - \Gamma_{\text{rel}} \cdot O_{\text{exc}} - \Gamma_{\text{OP}} \cdot O_{\text{exc}} - \Gamma_O \cdot O_{\text{exc}} - \Gamma_{\text{SF}} \cdot O_{\text{exc}}^2, \tag{43}
\]

\[
\frac{dO_T}{dt} = \Gamma_{\text{rel}} \cdot O_{\text{exc}} - \Gamma_{\text{OP}} \cdot O_T + \frac{1}{2} A_{\text{PP}} \cdot P_T^2 - \frac{3}{2} A_{\text{OO}} \cdot O_T^2 - \frac{1}{2} A_{\text{OP}} \cdot O_T \cdot P_T - \Gamma_O \cdot O_T - \Gamma_{\text{SF}} \cdot O_T^2, \tag{44}
\]

\[
\frac{dP_T}{dt} = \Gamma_{\text{OP}} \cdot P_T + \Gamma_{\text{OP}} \cdot O_{\text{exc}} - \frac{7}{4} A_{\text{PP}} \cdot P_T^2 - \Gamma_P \cdot P_T + \frac{1}{4} A_{\text{OO}} \cdot O_T^2 - \frac{3}{4} A_{\text{OP}} \cdot O_T \cdot P_T + \Gamma_{\text{SF}} \cdot O_{\text{exc}}^2 + \Gamma_{\text{SF}} \cdot O_T^2. \tag{45}
\]

The meaning of each symbol is given in table 2. We solved this set of equations numerically using the routines supplied by MATHCAD by giving the temporal shape of the laser pulse and the number of absorbed photons as initial values. The absorbed laser power was determined in each measurement by comparing the laser power ahead of and behind the cryostat windows and from the known reflection losses. For the data analysed in figure 9, the number of absorbed photons was determined from the absorbed laser power to be $N_{\text{abs}} = 6 \times 10^{10}$. The measured values agree well with those calculated from the indirect absorption process. The temperature was assumed to follow the dependence given by (40) with $T_{\text{in}} = 0.82$ K.

We found that the fit is not influenced at all by the spin–flip conversion, so we omitted this parameter in the final result. Also changing the lifetime of the orthoexciton in the reasonable range from 50 ns to 500 ns does not change the fit. We therefore set it equal to the paraexciton decay rate. Thus the only important parameters that sensitively changed the results of the rate equations are the paraexciton lifetime $1/\Gamma_P$ and the two-body decay constants $A_{\text{PP}}$, $A_{\text{OO}}$ and $A_{\text{OP}}$, whereby we assumed that $A_{\text{OP}} = (A_{\text{PP}} + A_{\text{OO}})/2$.  

New Journal of Physics  **14** (2012) 023054 ([http://www.njp.org/](http://www.njp.org/))
Figure 9. Fit of the temporal dependence of the exciton numbers by the rate model for orthoexcitons (A) and paraexcitons (B). Full symbols are the exciton numbers obtained from the spectral intensities according to the procedure described in the text; the crosses in (B) represent the exciton numbers obtained by assuming a constant conversion factor. The inset in (A) shows the dependence of the primarily excited orthoexcitons. The red points and line are taken with a gate width of 200 ns and the blue points and line with a gate width of 30 ns.

As the exciton numbers can change much faster than the gate time of our detector of 200 ns, we have to calculate from the solutions of the rate equations $O_{\text{exc}}(t)$, $O_T(t)$ and $P_T(t)$ corresponding averaged quantities defined by

$$\overline{N}_j(t) = \int_t^{t+T_G} N_j(t') \, dt',$$

which can be directly compared to the experiment. These functions are plotted in figure 9 for orthoexcitons in panel (A) and for paraexcitons in panel (B).

In the same way we can model the power dependence by integrating the results over the gate time of 200 ns and over 5 $\mu$s. The results of this calculation (full lines) are compared in figure 10 with the corresponding experimental data (symbols). In the analysis it turned out that the number of orthoexcitons in the trap during the excitation pulse is mostly determined by the two-body decay rate $A_{\text{OO}}$, which leads to the slight sublinear power dependence seen in figure 10(A). Both the lifetime of the paraexciton and its two-body decay rate have almost no influence on this dependence. From the analysis we obtain $A_{\text{OO}} = 1.7 \times 10^{-11} \text{ ns}^{-1}$. On the other hand, the number of orthoexcitons at later times (figure 10(B)) is determined almost completely by the regeneration from paraexcitons, i.e. by the paraexciton two-body decay rate and lifetime. To get agreement with the experimental data, we have to set $\Gamma_p = 1/650 \text{ ns}^{-1}$ and $A_{\text{pp}} = 1.7 \times 10^{-12} \text{ ns}^{-1}$, which is an order of magnitude smaller than $A_{\text{OO}}$.

As shown in figures 9 and 10, we obtain with these parameters very good agreement between the experimentally determined exciton numbers and the results of the rate model for both the temporal and the power dependence. The complete list of parameters used for these fits is given in table 1. It has to be stressed that, for the determination of the model parameters, the absolute number of paraexcitons is not needed, but comes out independently. Comparing with the paraexciton numbers obtained from fitting the spectra (full symbols in figures 9 and 10), we
Figure 10. Fit of the power dependence of the exciton numbers by the rate model. (A) Exciton numbers during the laser pulse (gate width 200 ns); (B) exciton numbers after the laser pulse (gate width 5 µs) as in figure 4. The inset in (A) shows the almost linear dependence of the number of excited excitons on the average laser power. The blue symbols and lines represent the orthoexcitons in the trap and the red symbols and lines the paraexcitons in the trap. Here the open and full diamonds are the numbers taken from the spectral analysis, while the crosses are obtained by assuming a constant conversion factor. The full and open symbols represent two experimental runs.

obtain excellent agreement, providing strong evidence of the correctness of our analysis. The paraexciton numbers in the power-dependent measurements can also be obtained quite well (but with more scatter in the data) using a constant conversion factor of $C = 1200$ from the intensities of the para ZP line, but this fails completely for the time-dependent results, which shows that the assumption in [24] of a constant conversion factor is highly questionable.

The values for the volume Auger rates calculated from the trap rates $A_{0\text{OO}}$ and $A_{0\text{PP}}$ are given in table 1. The value for the orthoexciton rate agrees very well with that in [16], while that for the paraexciton is a factor of 5 lower (see table I in [24]). In view of the uncertainty stated in this paper for their value of the paraexciton rate at low stress, which they could barely observe at all, we think that our results are more reliable. All the other investigations on the two-body decay of excitons, which gave much higher values for the two-body decay rates, have been performed in unstrained samples [18, 19]. It seems that in an unstrained sample the excitons behave quite differently and thus both experiments are not directly comparable. This might be related to the different orthoexciton spin states in the trap, since the state $|xy\rangle$ is pushed to higher energies and is no longer involved in the scattering processes.

6. The possibility of a Bose–Einstein condensation

The experiments presented have shown that under pulsed excitation it is possible to collect more than $2 \times 10^{10}$ paraexcitons in a trap with an initial effective exciton temperature of the order of 10 K, which is too high for a BEC to occur. For times later than 1000 ns, the excitons have
Figure 11. Modelling the paraexciton transition to a Bose–Einstein condensate for different temperatures of the He bath as indicated by $T_{\text{fin}}$. The blue solid line marks the exciton number, the dotted line the exciton temperature $T$ and the red solid line the condensate fraction $\eta_{\text{BEC}}$. The number of initially excited excitons was set to $2 \times 10^{10}$. In order to stabilize the condensate, we assumed in the rate equations that the excitons in the condensate do not underlie the two-body decay process.

cooled down to the bath temperature, but here the number of excitons in the trap has fallen down to below $10^8$, which again is too small for a BEC to occur at our bath temperature of 820 mK. However, the rate model allows us to answer the question of whether the transition to a BEC is possible at all by adjusting the external conditions such as bath temperature and laser pumping. In a first step we solved the set of rate equations to obtain the number of paraexcitons in the trap $N(t) = P_T(t)$ under the parameters derived from the experiment assuming different conditions concerning the bath temperature. Here we assume that the cooling of the excitons still follows (40) with the final temperature given by the bath temperature ($T_{\text{fin}} = T_{\text{bath}}$). BEC of the excitons that are assumed to behave like an ideal Bose gas occurs if for a given set $N(t)$, $T(t)$ the critical condition $k_B T_C = \hbar \Omega_0 (N_C / \zeta(3))^{1/3}$ (compare (1)) is fulfilled. The condensate fraction $\eta_{\text{BEC}}$ is given by $\eta_{\text{BEC}}(t) = 1 - N_C / N(t)$. The results shown in figure 11, where the number of paraexcitons and condensate fraction are plotted by blue and red lines, respectively, demonstrate that one way towards a BEC in our specific sample would consist in a dramatic lowering of the bath temperature down to 50 mK. In principle, this seems to be possible in our mixing cryostat by further minimizing the heat input through the windows. Another promising way to achieve BEC would consist in lowering the initial exciton temperature by changing the excitation pathway. The reason for the high initial temperature in our experiment is certainly the large kinetic energy excess of the paraexcitons after conversion from the orthoexcitons, which
would correspond to an effective temperature of about 60 K if relaxation by phonon scattering did not occur. We therefore propose to use direct excitation of paraexcitons via process III in figure 3(A). Here the temperature of the paraexcitons is determined by the difference between the laser photon energy and the energy of the trap minimum, which can be reduced down to zero. However, this excitation process implicates severe experimental problems: one problem is the weakness of this absorption, which requires high laser power to achieve the necessary exciton numbers. Another problem is the scattered laser light, which is spectrally very close to the paraexciton luminescence and outshines it by several orders of magnitude. This might be overcome by using appropriate spatial and polarization filtering of the detected light.

7. Conclusions and outlook

We have presented experiments on the relaxation dynamics of para- and orthoexcitons in Cu$_2$O at bath temperatures down to 820 mK in a confining potential of several meV depth. These show, in contrast to previous measurements in bulk material and in agreement with measurements in potential traps at bath temperatures above 2 K, that the two-body decay process is drastically reduced and of the order of $10^{-18}$ cm$^3$ ns$^{-1}$. While in the present experiments due to the high temperature of the excitons no transition to a Bose–Einstein condensate could be observed, we suggest two scenarios which might allow this observation. These require to reduce the bath temperature down to $<100$ mK and, most effectively, the reduction of the initial paraexciton temperature. Experiments for realizing these scenarios are under way.

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New Journal of Physics 14 (2012) 023054 (http://www.njp.org/)