# Many-Body Dynamics and Exciton Formation Studied by Time-Resolved Photoluminescence

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The dynamics of exciton and electron-hole plasma populations is studied via time-resolved photoluminescence after nonresonant excitation. By comparing the peak emission at the exciton resonance with the emission of the continuum, it is possible to experimentally identify regimes where the emission originates predominantly from exciton and/or plasma populations. The results are supported by a microscopic theory which allows one to extract the fraction of bright excitons as a function of time.

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## I. INTRODUCTION

Excitons, i.e. bound states between a conduction band electron and a valence band hole, are often regarded as the semiconductor analogue of bound hydrogen atoms. In contrast to the hydrogen atom, however, the exciton binding energy lies in the range of 10 meV due to the small masses and the large background dielectric constant<sup>1</sup>. Furthermore, typical excitation conditions in semiconductors lead to phase-space filling and screening such that for elevated carrier densities the Coulomb interaction becomes screened and the binding even weaker. Thus, only at low lattice temperatures and up to moderate densities can bound exciton populations be expected to be the dominating species in the interacting electron-hole system. In general, the many-body state is a complicated mixture containing Coulomb-correlated electrons and holes as well as bound and ionized exciton correlations and populations.

Even more importantly, under typical experimental situations the real hydrogen atom in its ground state is stable against decay while excitons first have to be generated by optical excitation and subsequent many-body interactions, and then they can recombine radiatively or nonradiatively. It is known that the radiative lifetime in low-dimensional systems is usually much shorter than in the respective bulk materials<sup>2,3</sup>. Consequently, after nonresonant excitation where the laser is tuned to energies corresponding to the continuum of electron-hole transitions, it is interesting to find out if and how many excitons form and which states they occupy. The formation process involves Coulomb scattering and interaction with lattice vibrations (phonons). Under favorable conditions, electron-hole pairs can give their excess energy to the phonon bath or to the remaining carrier system and form bound excitons<sup>4</sup>. Since the photon momentum is small compared to typical exciton momenta, only excitons with extremely small momenta, i.e. inside the light cone, can radiatively recombine. We refer to them as *bright excitons*. The remaining *dark excitons* first have to scatter down into the light cone before they can decay radiatively. Hence, one would generally assume that under nonresonant excitation conditions with subsequent exciton formation the vast majority of these bound pairs would always exist in the different dark states.

The finite radiative lifetime of bright excitons is on the order of 13 ps at low temperatures in GaAs based single quantum-well systems. Monitoring the time-dependent photoluminescence (PL) collected at the 1s-exciton resonance therefore yields information about the dynamics of these exciton populations. The build-up of the PL after nonresonant excitation has been interpreted as build-up of an incoherent exciton population<sup>5,6,7,8,9,10</sup>, and the subsequent fall off was taken as evidence for this population decay<sup>2,11</sup>. However, a recently developed microscopic luminescence theory for a Coulomb correlated plasma of electron-hole pairs predicts that luminescence peaks at the 1s-exciton resonance even without the presence of incoherent exciton populations<sup>12</sup>. In fact, the cooling dynamics of an unbound electron-hole plasma after interband excitation leads to a luminescence spectrum whose peak grows and sharpens at the 1s-exciton resonance as the carriers relax to the bottom of their respective bands. Accordingly, the mere existence of PL at the spectral position of the 1s-resonance would not prove the existence of an incoherent population of excitons, and previous interpretations using a purely excitonic picture need to be reviewed carefully.

Since the energy differences between exciton states correspond to frequencies in the terahertz regime, terahertz

absorption measurements provide a powerful alternative method to study exciton formation<sup>13,14</sup>. Theoretically, it has been shown that the build-up of the induced absorption corresponding to the exciton 1s to 2p transition is sensitive only to the correlated exciton populations and gives thus direct evidence for exciton populations<sup>15,16</sup>. Current results from terahertz absorption measurements after nonresonant excitation suggest exciton formation times of several hundreds of picoseconds<sup>13</sup>.

A recent publication based on photoluminescence measurements combined with a microscopic theoretical model for emission from plasma and excitons<sup>17</sup> shows that only a small fraction of the carrier density is transformed into optically active excitons under best formation conditions. However, even this small bright 1s-exciton population can lead to massive changes of the emission spectra. Clearly, since the PL is not sensitive to dark excitons, these measurements do not allow to unambiguously determine the total number of excitons in the system. Nevertheless, it seems to be established by now that under suitable density and excitation conditions an exciton population is present at a liquid-helium lattice temperature ( $\approx 4 \text{ K}$ ) and that this population may dominate the 1s-emission properties. The precise formation dynamics and in particular the interpretation of time resolved PL measurements, however, is still controversial. For example, a recent experiment<sup>18</sup> shows that in nonresonantly excited time resolved PL measurements the 1s-resonance is developed on a sub-ps timescale at 100 K, much faster than any expected exciton formation time. We interpret this experiment as direct support for the predicted emission at the 1s-resonance originating from populations of unbound, Coulomb correlated electron-hole plasma states.

In the present work, we complement our published experimental results<sup>17</sup> by a detailed study of time-resolved PL combined with nonlinear absorption measurements to determine the carrier density. After an outline of the experimental and theoretical methods in Sec. II we discuss our results in Sec. III. We show that from the investigation of the dynamics of the experimental PL spectra alone, one can clearly distinguish two regimes with and without major exciton contributions. This confirms our previous result that above 30 K the emission at the 1s-resonance is dominantly due to unbound pair-state emission, while for temperatures lower than 30 K an increasing optically active exciton population contributes<sup>17</sup>. We support our experimental results by a microscopic theory leading to the luminescence analogue of the famous Elliott formula<sup>19</sup>. It includes not only the exciton resonances but also a source term which contains both a plasma contribution and an incoherent exciton population. This theory is used to extract a time resolved number of bright excitons.

### II. METHODS

#### A. Experiment

The principal sample (DBR42) consists of twenty  $In_{0.06}Ga_{0.94}As$  quantum wells. Each is 8 nm thick and grown in between 130 nm GaAs barriers. The indium concentration and well thickness were chosen to place the 1s heavy-hole (hh) exciton resonance at 1.471 eV at 4 K lattice temperature. The linewidth of the exciton transition is 0.96 meV full width at half maximum (FWHM); the exciton binding energy is 8 meV. This sample permits the detection of about 13 meV of emission above the 1s hh exciton resonance, undistorted by substrate absorption and impurities. At the same time, the 1s-transition energy is high enough to lie within the operating range of laser and detector. We excited nonresonantly 13.2 meV above the 1s-resonance, into the heavy-hole continuum but below the light-hole resonance. Both sides of the sample were anti-reflection coated to reduce Fabry-Pérot interference fringes. The experimental results were checked on several other samples.

Photoluminescence and nonlinear absorption measurements were performed under identical conditions, i.e. sample temperature and excitation density. The linear and nonlinear absorption of the sample under investigation was measured with a ps pump-probe setup. Both pulses were generated by a solid-state pumped, actively mode-locked Titanium:Sapphire laser with a fixed repetition rate of 80 MHz and a maximum time-integrated output power of 2 W. The peak wavelength of the spectrally broad 100 fs probe pulse, tunable using a birefringent filter, was typically centered at the heavy-hole continuum edge.

The long PL lifetime of several nanoseconds required a reduction of the laser repetition rate. For the experiments presented here, an electro-optic modulator reduced it to 2 MHz to eliminate carrier accumulation effects in the sample. To measure the absorption corresponding to PL signals that take 11 ns to decay, it was necessary to expand and collimate the probe beam and pass it twice through a  $\approx 1$  m delay line. For selective excitation, a spectrally narrower tunable 3 ps pump pulse was generated in a grating pulse shaper. The spectral width of the pump pulse was 2 meV FWHM. The sample was held in a cold finger cryostat. The pump and probe pulses were focussed onto the sample using 10 cm and 45 cm focal length lenses, yielding 60  $\mu$ m and 20  $\mu$ m spot sizes, respectively. The transmission and PL were spectrally resolved using identical imaging grating monochromators with a spectral resolution of 0.8 meV. The transmitted probe light was detected using a liquid nitrogen cooled charged-coupled device (CCD) camera, and the PL with a Hamamatsu streak camera with a slow sweep unit in single photon counting mode. This mode of operation

reduces the time resolution to 90 ps. To even better utilize the dynamic range of the streak camera, a film neutral density filter was placed at the exit plane of the monochromator reducing the fluence by a factor of 40 for all energies  $< 1.474 \,\mathrm{eV}$ . This was extremely helpful because a partial goal of this study was to detect several meV of continuum emission which is intrinsically four to five orders of magnitude lower than the emission peak at the 1s-resonance. All data shown here have been corrected for this attenuation and are presented undistorted. Photoluminescence spectra are integrated from t-50 ps to t+50 ps around each time step.

To estimate the carrier density  $n_{\rm eh}$  during the decay at time t after the pump pulse,  $n_{\rm eh}(t)$  is assumed to be linearly proportional to the change in absorption  $\Delta \alpha L$  at time t. From the absorption at the pump energy, the measured pump power and the pump spot size at the sample, the initial carrier density was estimated and the corresponding nonlinear absorption was measured at 10 ps before any reduction occurred. This calibration curve linking  $\Delta \alpha L$  and  $n_{\rm eh}$  was used to determine the carrier density at any time delay. This calibration might not be completely unambiguous, since the nonlinear absorption spectrum is influenced differently by carriers bound to excitons,  $n_X$ , as compared to quasi-free carriers,  $n_{\rm eh} - n_X$ . However, since in the following experiments in general  $n_X \ll n_{\rm eh}$ , we neglect the subtle differences and believe that the nonlinear absorption still provides an accurate measure of the total carrier density.

### B. Theory

In order to interpret the experimental observations quantitatively, we apply our microscopic quantum theory. In the current approach we utilize findings from previous publications<sup>4,17</sup> which allow us to compute the PL spectrum under quasi steady-state conditions and to clearly distinguish between bound and unbound pair state contributions. The theory starts from the fundamental Hamiltonian including the quantized carriers coupled via Coulomb interaction, and interacting with phonons and a quantized light field. This theory, evaluated at the level of a Hartree-Fock approximation, first predicted PL at the exciton energy without exciton populations<sup>12</sup>. Meanwhile we have extended the analysis to include also electron-hole correlations and bound excitons<sup>4,15</sup>.

The fundamental quantity for the computation of a PL spectrum is the rate of emitted photons,

$$I_{\rm PL}(\omega_q) = \frac{\partial}{\partial t} \Delta \langle B_q^{\dagger} B_q \rangle = \frac{2}{\hbar} \operatorname{Re} \left[ \sum_k \mathcal{F}_q^{\star} \Delta \langle B_q^{\dagger} a_{v,k}^{\dagger} a_{c,k} \rangle \right], \tag{1}$$

where  $B_q^{\dagger}B_q$  is the photon number operator for photons with wavenumber q,  $a_{\lambda,k}^{\dagger}$  creates an electron in band  $\lambda = c, v$  in quantum state k, and  $\mathcal{F}_q$  denotes the coupling matrix element to mode q. We have restricted the analysis to emission perpendicular to the quantum well such that the wave vector q is a scalar quantity and equivalent to the frequency  $\omega_q = c_0 q$ . In Eq. (1), the  $\Delta \langle \ldots \rangle$  denotes the correlated (i.e. incoherent) part and is given by the full expectation value minus its factorized (i.e. classical) contribution<sup>20,21</sup>.

The photon number and thus the PL are coupled to photon-assisted polarizations of the form  $\Delta \langle B^{\dagger} a_{v}^{\dagger} a_{c} \rangle$ . Therefore, we set up the equation of motion for this quantity,

$$\frac{\partial}{\partial t} \Delta \langle B_{q}^{\dagger} a_{v,k}^{\dagger} a_{c,k} \rangle = (\epsilon_{k} - \hbar \omega_{q} - i \gamma_{k}^{\mathrm{D}}(\omega_{q})) \Delta \langle B_{q}^{\dagger} a_{v,k}^{\dagger} a_{c,k} \rangle - (1 - f_{k}^{e} - f_{k}^{h}) \sum_{k'} V_{k-k'} \Delta \langle B_{q}^{\dagger} a_{v,k'}^{\dagger} a_{c,k'} \rangle$$

$$+ i \sum_{k'} \gamma_{k,k'}^{\mathrm{OD}}(\omega_{q}) \Delta \langle B_{q}^{\dagger} a_{v,k'}^{\dagger} a_{c,k'} \rangle + i \mathcal{F}_{q} (f_{k}^{e} f_{k}^{h} + \Delta \langle a_{c,k'}^{\dagger} a_{v,k}^{\dagger} a_{c,k} a_{v,k'} \rangle), \qquad (2)$$

where we have neglected a stimulated contribution which can describe radiative coupling between multiple quantum wells and is not important under the experimental conditions. Here,  $\epsilon_k$  is the total kinetic energy of electron and hole in state k, and  $f_k^{e(h)}$  describes the microscopic carrier distribution of electrons (holes). Furthermore, we have included Coulomb scattering via  $\gamma_{k,k'}^{OD}(\omega_q)$  and  $\gamma_k^D(\omega_q)$ . These scattering matrices are obtained from solving and inserting the higher order correlations  $\Delta \langle B^{\dagger} a_v^{\dagger} a_{\lambda}^{\dagger} a_{\lambda} a_c \rangle$  in second-Born approximation. The last term of Eq. (2) provides the source for spontaneous emission. It contains the Hartree-Fock contribution proportional to the product of electron and hole distributions<sup>12</sup> and also a correlated source. This correlated source is a true two-particle expectation value and changes depending on whether electrons and holes form a correlated plasma or whether additional bound excitons are present. This term will be discussed in detail below.

In order to calculate the PL spectrum, we use an adiabatic treatment for the photon-assisted polarizations and introduce a generalized Wannier basis which diagonalizes the homogeneous part of Eq. (2). Due to the phase-space filling factor  $(1 - f_k^e - f_k^h)$  and the presence of the second-Born scattering matrices, the respective matrix is not Hermitian and we have to distinguish between right- and left-handed eigenfunctions  $\phi_{\nu,q}^{l/r}$ . Furthermore, we obtain complex eigenvalues of the form  $E_{\nu}(\omega_q) - i\gamma_{\nu}(\omega_q)$ . Both real and imaginary part of these eigenvalues depend on the photon energy through the energy denominator in the second-Born scattering rates. Since the imaginary part of the eigenvalue depends on the excitonic state, this basis also correctly describes the fact that energetically higher lying exciton states show nonlinear behavior in the form of a larger linewidth for much lower densities than the lowest bound state. Using this exciton basis, we obtain the steady-state PL as

$$I_{\rm PL}(\omega_q) = \frac{|d_{\rm cv}^2|\omega_q}{\varepsilon_{\rm bg}} {\rm Im} \Big[ \sum_{\nu} \frac{\phi_{\nu,q}^r(r=0)}{E_{\nu}(\omega_q) - \hbar\omega_q - i\gamma_{\nu}(\omega_q)} \sum_{k,k'} [\phi_{\nu,q}^l(k)]^* \langle a_{c,k'}^\dagger a_{v,k'} a_{v,k'}^\dagger a_{c,k'}^\dagger \Big].$$
(3)

The prefactor in Eq. (3) is determined by the square of the dipole matrix element  $|d_{cv}|^2$  and the background dielectric constant  $\varepsilon_{bg}^{22}$ . Equation (3) is reminiscent of the famous Elliott formula for bandgap absorption<sup>19</sup>; it contains a sum over exciton states, and the resonances of the denominator show that the PL peaks at the same excitonic energies as the absorption. In fact, a solution of the nonlinear susceptibility similar to the method above leads to

$$\chi(\omega_q) = \frac{|d_{\rm cv}^2|}{\varepsilon_{\rm bg}} {\rm Im} \Big[ \sum_{\nu} \frac{\phi_{\nu,q}^r(r=0)}{E_{\nu}(\omega_q) - \hbar \omega_q - i \gamma_{\nu}(\omega_q)} \sum_k [\phi_{\nu,q}^l(k)]^* \left(1 - f_k^e - f_k^h\right) \Big],\tag{4}$$

with the same excitonic basis  $\phi_{\nu,q}$ , energies  $E_{\nu}(\omega_q)$ , and damping constants  $\gamma_{\nu}(\omega_q)$ . Due to the frequency dependence of the  $\gamma_{\nu}(\omega_q)$ , Eqs. (3) and (4) can describe non-Lorentzian lineshapes. In the limit of low densities, the  $\gamma_{\nu}$  approach zero and the phase-space filling factor becomes approximately 1, and we recover the low-density Elliott formula. In contrast to the absorption, the strength of the PL according to Eq. (3) is not only determined by the exciton wavefunctions but also by the source term  $\sum_{k,k'} [\phi_{\nu,q}^l(k)]^* \langle a_{c,k'}^{\dagger} a_{c,k} a_{v,k'} a_{v,k}^{\dagger} \rangle$ . This source contains a factorized contribution

$$\sum_{k} [\phi_{\nu,q}^{l}(k)]^{*} f_{k}^{e} f_{k}^{h} = \sum_{\nu'} [\phi_{\nu',q}^{r}(r=0)]^{*} \langle X_{\nu',q}^{\dagger} X_{\nu,q} \rangle_{\mathrm{HF}}$$
(5)

with

$$\langle X_{\nu',q}^{\dagger} X_{\nu,q} \rangle_{\rm HF} = \sum_{k} \phi_{\nu',q}^{l}(k) \, [\phi_{\nu,q}^{l}(k)]^{\star} f_{k}^{e} f_{k}^{h}, \tag{6}$$

which is always present as soon as electrons and holes are excited. This is the source term due to which 1s-luminescence without exciton populations has been predicted first<sup>12</sup>. Additionally, the correlated part of the source can describe a correlated plasma as well as additional incoherent bound exciton correlations  $N_{1s}$  which may or may not be in the system. From our previous results<sup>4</sup> we know that even without the formation of any bound excitons, the two-particle correlations do not vanish and describe a correlated plasma. Without the inclusion of the second-Born scattering discussed above, it can be shown that the presence of the correlations mainly cancels all the off-diagonal contributions  $\nu \neq \nu'$  in Eq. (5). In the present paper, however, we numerically solve the full sum of Eq. (5) and treat the correlated part explicitly.

Thus, the last missing input for the evaluation of Eq. (3) is the two-particle correlations. The full equation of motion without Coulomb scattering has been given in a previous publication<sup>4</sup>. For the present theory-experiment comparison, we also include second-Born scattering which is provided by an approximative solution of six-point quantities. Similar to the photon-assisted quantities, the equation of motion for the exciton correlations contains a homogeneous part and an external source depending on the carrier distributions  $f^e$  and  $f^h$ . We therefore proceed similarly as with the photon-assisted correlations in that we solve the homogeneous equation by transforming into an adapted exciton basis  $\tilde{\phi}_{\nu}^{l/r}$ . This basis set is different from the one used for the photon-assisted polarizations because for the pure particle correlations there is no dependence on the optical frequency  $\omega_q$  such that the second-Born scattering rates and consequently the complex eigenvalues  $\tilde{E}_{\nu} - i\tilde{\gamma}_{\nu}$  are different from the ones discussed above. Using this new basis we can express the correlated source as

$$\sum_{k,k'} [\phi_{\nu,q}^{l}(k)]^{*} \Delta \langle a_{c,k'}^{\dagger} a_{v,k'} a_{v,k}^{\dagger} a_{c,k} \rangle = \sum_{\nu',\nu''} [\tilde{\phi}_{\nu'}^{r}(r=0)]^{*} \left( \sum_{k} [\phi_{\nu,q}^{l}(k)]^{*} \tilde{\phi}_{\nu''}^{r}(k) \right) \Delta \langle X_{\nu'}^{\dagger} X_{\nu''} \rangle$$
(7)

where  $X_{\nu}$  is the exciton annihilation operator. The term  $\Delta \langle X_{\nu'}^{\dagger} X_{\nu''} \rangle$  can easily be obtained from the adiabatic solution of the Heisenberg equation of motion. In the absence of phonon scattering or other mechanisms leading to exciton formation it is given by

$$\Delta \langle X_{\nu'}^{\dagger} X_{\nu''} \rangle_{\rm pl} = -\frac{S_{\nu',\nu''}}{\tilde{E}_{\nu''} - \tilde{E}_{\nu'} - i(\tilde{\gamma}_{\nu''} + \tilde{\gamma}_{\nu'})}$$
(8)

with the single particle source  $S_{\nu',\nu''}$  given by

$$S_{\nu',\nu''} = \sum_{k,k'} \tilde{\phi}_{\nu'}(k') [\tilde{\phi}_{\nu''}(k'')]^* V_{k'-k''} \left[ (1 - f_{k'}^e - f_{k'}^h) f_{k''}^e f_{k''}^h - (1 - f_{k''}^e - f_{k''}^h) f_{k'}^e f_{k'}^h \right].$$
(9)

The proper inclusion of Coulomb scattering and the distinction between the different basis sets for photon-assisted polarizations and correlations, respectively, is vital for a quantitative theory-experiment comparison over the experimentally relevant density regime. Everything mentioned so far is regarded by us as PL resulting purely from unbound pair states since in the end all terms depend solely on single-particle electron and hole distribution functions. For the comparison with the experiment, we take the experimentally estimated carrier density and temperature as input for these single-particle distributions and calculate the emission. In addition, e.g. in the presence of phonon scattering, extra bound 1s-excitons may form as discussed in Ref. 4. In the present paper, we do not include the formation dynamics explicitly; instead, we add a purely exciton contribution  $N_{1s,q=0}$ 

$$\Delta \langle X_{\nu'}^{\dagger} X_{\nu''} \rangle = \Delta \langle X_{\nu'}^{\dagger} X_{\nu''} \rangle_{\text{pl}} + \delta_{\nu',1\text{s}} \delta_{\nu'',1\text{s}} N_{1s,q=0}$$
(10)

of optically active 1s-exciton correlations in Eq. (7). The exciton contribution is thus treated as input to the theory and varied until agreement with the experimental spectra is reached. Thereby, we can distinguish between bound and unbound pair-state populations and extract the number of optically active ( $q \approx 0$ ) 1s-excitons.

# III. RESULTS

#### A. Correlated plasma regime

Nonlinear absorption measurements (left) and corresponding PL spectra (middle and right) are shown in Fig. 1 at a time delay of 1 ns after excitation. The initial carrier density is  $3.3 \times 10^9$  cm<sup>-2</sup>, and the nominal lattice temperatures are (a) 70 K, (b) 50 K and (c) 30 K. The absorption spectra (left) exhibit a strong absorption at the 1s-exciton resonance followed by a 2s peak and the onset of the electron-hole continuum. To allow for easier comparison of the spectral shape, the energy scale is chosen relative to the 1s-exciton absorption peak at 1.4650 eV (70 K), 1.4681 eV (50 K), and 1.4697 eV (30 K). The line broadens only slightly in this temperature range<sup>23</sup>.

The corresponding PL spectra are shown on a linear scale (middle) and on a semi-logarithmic scale (right). The emission is strongly peaked at the 1s-exciton resonance and clearly dominates the linear luminescence spectra. The 2s emission is seen as well. A small discontinuity on the high energy slope between 3 meV and 7 meV above the 1s peak is visible in the PL spectra; it is due to the imperfect correction of the attenuator used for lower energies. While the main features of the emission stay the same, namely that the spectrum is dominated by a peak at the 1s-resonance for all shown lattice temperatures, the most obvious difference is the decreasing slope of the continuum emission with increasing temperatures.

Coulomb scattering causes the electrons and holes to thermalize to Fermi-Dirac distributions with a carrier temperature T, often much higher than the lattice temperature  $T_L$ , on a sub-picosecond time scale<sup>24,25</sup>. Therefore, within the achieved time resolution, thermalization can be regarded as instantaneous. To investigate the carrier temperature further we assume that the electrons and holes in the continuum are in thermal equilibrium. In thermodynamic equilibrium, the Kubo-Martin-Schwinger (KMS) relation<sup>26,27</sup>

$$I_{\rm PL}^{\rm eq}(\hbar\omega) \propto g(\hbar\omega - \mu)\,\alpha(\hbar\omega) \tag{11}$$

is sometimes used to establish the relation between the absorption coefficient  $\alpha$  and the PL. Here,  $\mu$  is the joint chemical potential of the electrons and holes, and  $g(E) = (\exp(\frac{E}{k_BT}) - 1)^{-1}$  is the Bose distribution function. For sufficiently low densities, the Bose function can be approximated by a Boltzmann distribution function  $g(E) \approx A \exp(-\frac{E}{k_BT})$ . This approximation is justified for the density and temperature range investigated here. Thus the emission decreases exponentially above the band edge, and the carrier temperature can be extracted from the slope of the continuum emission. Due to experimental noise, a small spectrally and temporally flat background may be present in the data; it is subtracted in all data shown. The data are therefore fit using a least mean square fit for the variables, amplitude A and carrier temperature T, with the corresponding standard deviations  $\sigma_A$  and  $\sigma_T$ . Figures 2(d), (e), and (f) show the temperatures extracted from the PL data under the conditions of Fig. 1 that displays the 1 ns spectra. The carrier temperatures approach values close to the lattice temperature in all three cases. Times  $\leq 0.1$  ns are not accessible because of limited time resolution of the experiment; also the concept of a carrier temperature is ambiguous because of highly nonequilibrium Coulomb scattering processes at early times.



FIG. 1: Nonlinear absorption spectra (left) and PL spectra on a linear scale (middle) and logarithmic scale (right) for a time delay of 1 ns for various lattice temperatures (a) 70 K, (b) 50 K, and (c) 30 K. The initial carrier density is  $n_{\rm eh} = 3.3 \times 10^9 \, {\rm cm}^{-2}$ .

The rise and subsequent decay of the PL were detected time-resolved. The time evolution of the 1s-emission can be studied in several ways. It is possible to spectrally integrate between the FWHM points or all energies below the midpoint between 1s and 2s. Alternatively, the peak of the emission spectrum can be traced. Here the spectrum is integrated  $50 \,\mu\text{eV}$  around the peak of the linear absorption spectrum. Use of any of the other methods of analysis does not change the results significantly. The continuum emission is spectrally integrated from the band edge to  $100 \,\mu\text{eV}$ above it to achieve an acceptable signal to noise ratio. A representative set of normalized emission spectra can be seen in Fig. 2 (a)–(c) for nominal lattice temperatures of 70 K (a), 50 K (b), and 30 K (c), shown on a semi-logarithmic scale. Compared within each plot are the emission from the 1s-exciton resonance (open triangles) and the emission from the continuum edge (full squares).

Clearly, the emission from the 1s-exciton resonance and the continuum emission have the same temporal dependence for lattice temperatures of 70 K and 50 K, strongly indicating that the emission source is the same in both cases. The peak of the continuum and 1s PL is reached shortly after the end of the excitation. For a lattice temperature of 30 K, 1s-emission and continuum emission start to deviate slightly: The emission from the 1s-exciton resonance continues to increase slightly after the end of the excitation pulse. This deviation becomes more and more pronounced for lower lattice temperatures, as discussed in the next section.

To investigate the time evolution of the PL further for situations where exciton populations can be omitted, we apply a simple rate equation model. Under such conditions, the Elliott formula Eq. (3) predicts a plasma source term  $S_{\rm pl} \propto n_{\rm eh}^2(t)$  in the low density limit. Therefore, the continuum emission reduces to the well known  $I_{PL} \propto B n_{\rm eh}^2(t)$ , where B is the electron-hole bimolecular recombination rate. Accordingly, the temporal evolution of the carrier density is governed by the bimolecular recombination rate:  $dn_{\rm eh}/dt = -B n_{\rm eh}^2$ . However, the PL in the experimentally investigated time range cannot be fitted by only using the radiative decay proportional to B; a nonradiative recombination  $\propto -A n_{\rm eh}$  must be included, which describes the exponential like decay at longer times. As the temperature is increased, the nonradiative decay becomes faster, which can be at least partially explained by carriers escaping from the quantum well; especially the holes escape easily due to their shallow quantum well. Subsequently, carriers may decay nonradiatively and emit PL elsewhere.



FIG. 2: Temporal evolution of the PL of the 1s-exciton resonance (open triangles) and of the continuum edge (full squares) for an initial carrier density of  $n_{\rm eh} = 3.3 \times 10^9 \, {\rm cm}^{-2}$  at lattice temperatures of (a) 70 K, (b) 50 K, and (c) 30 K. Fits using the simplified plasma model are shown as solid lines. The corresponding temporal evolution of the carrier temperature is shown on the right side at lattice temperatures of 70 K (d), 50 K (e) and 30 K (f).

The corresponding fits using the simplified model without exciton populations are included in Fig. 2 (a)–(c) as solid lines. As can be seen in the figure, the simplified model describes well the experimental data. The radiative recombination coefficient B agrees within a factor of 2 to 3 with the expression given in Ref. 28. Hence, the coinciding temporal evolution of the 1s and the continuum emission together with the well fitting model unambiguously point to the unbound electron-hole plasma as the sole source of emission from both the 1s-exciton resonance and the electron-hole continuum for lattice temperatures T > 30 K.

### B. Excitonic regime

Lowering of the lattice temperature further leads to a changing picture. At 4K lattice temperature, the time evolution of the emission from the 1s-exciton resonance differs significantly from that of the continuum: The maximum of the peak 1s-emission is delayed compared to that of the continuum emission; see Fig. 3. For lower excitation densities, the maximum is reached about 0.8 ns after excitation. With increasing carrier densities, the maximum occurs earlier. For the highest investigated density the luminescence maximum is reached after 0.4 ns. Thus, the 1s PL develops its own dynamics and cannot be solely described by emission from unbound carriers; luminescence from an incoherent exciton population also contributes<sup>17</sup>.

To further analyze and quantify the amount of bright excitons contributing to the 1s PL, we introduce a single parameter  $\beta^{17,29}$ , defined as

$$\beta = \frac{I_{\rm PL}(1s)}{I_{\rm PL}^{\rm eq}(1s)}.\tag{12}$$

Here,  $I_{\rm PL}(1s)$  is the measured PL at the 1s-resonance or the calculated one by applying our microscopic theory.  $I_{\rm PL}^{\rm eq}(1s)$  satisfies the KMS relation Eq. (11). Experimentally and theoretically,  $I_{\rm PL}^{\rm eq}(1s)$  is found by multiplying the



FIG. 3: Temporal evolution of the PL of the 1s-exciton resonance (open triangles) and of the continuum edge (full squares) for an initial carrier density of  $n_{\rm eh} = 3.3 \times 10^9 \,{\rm cm}^{-2}$  at a lattice temperature of 4 K. The fit using the simplified plasma model is shown as solid line.

measured and calculated nonlinear  $\alpha$  by a Boltzmann factor, using the temperature extracted from the measured continuum emission, and normalizing it to agree with the measured and calculated continuum PL. Thus,  $\beta$  quantifies how the 1s-emission of a given spectrum differs from that expected from the measured absorption assuming validity of the KMS relation, with the measured and KMS PL spectra forced to agree in the continuum.

The theory-experiment comparison is shown in Fig. 4 for an initial carrier density of  $1.9 \times 10^{10}$  cm<sup>-2</sup>. The temporal evolution of the experimentally deduced carrier density is plotted in Fig. 4 (b). Figure 4 (a) shows the temporal evolution of the carrier temperature extracted from the continuum tail of the corresponding measured luminescence spectra (full squares). For all investigated carrier densities the electron-hole plasma cools exponentially with typical decay times of 0.5 to 0.6 ns . However, in contrast to the data series taken at higher lattice temperatures, the carrier temperature never reaches the lattice value of 4 K<sup>29,30,31,32</sup>.

The input parameters to our theoretical analysis are the carrier temperature and density. While the density can directly be taken from the experiment, the extracted temperature from the tail of the continuum PL is not necessarily identical to the electronic temperature which enters the theory. Our theory shows that as a consequence of Coulomb scattering the extracted temperature from the continuum emission is always slightly higher than the carrier temperature put into the simulation. Therefore, our analysis has to be done self-consistently and we have run simulations with carrier temperatures adjusted in such a way that the resulting continuum tail of the spectrum agrees with the experiment. The carrier temperatures put into the simulations are shown as full triangles in Fig. 4 (a). We note that especially for early times with relatively high carrier density and corresponding stronger scattering the deviation between carrier temperature and extracted continuum slope is appreciable and stronger than for later times.

Figure 4 (c) displays the time dependence of  $\beta$ . Experimental values are plotted as full squares. The corresponding theoretical values of the bare plasma emission are shown as full triangles. They decrease as time goes by as a consequence of the decreasing carrier density. This is in full agreement with our previous findings. As discussed in Ref. 17, the calculation without exciton populations underestimates the measured  $\beta$  and an optically active q = 0 1s-exciton population has to be added, which strongly enhances the 1s PL. Excitons are added until the theoretical and experimental  $\beta$ 's are equal<sup>33</sup>; see open triangles in Fig. 4 (c).

Since PL and thus  $\beta$  are only sensitive to bright excitons, only the number  $N_{1s,q=0}$  of 1s-excitons with vanishing center-of-mass momentum can be directly obtained from our theory. In order to extract the bright exciton density



FIG. 4: (a) Temperature dynamics as extracted from the experimental PL spectra (squares) and carrier temperatures used as input for the theory (triangles). The lattice temperature is 4 K. (b) Temporal evolution of the experimentally deduced carrier density. The initial carrier density is  $n_{\rm eh} = 1.9 \times 10^{10} \text{ cm}^{-2}$ . (c) Parameter  $\beta$  as function of time: experimentally (full squares), calculated with (open triangles) and without (full triangles) inclusion of an exciton population. (d) Extracted fraction of bright excitons.

 $n_{\rm X}^{\rm br}$  from  $N_{1{\rm s},q=0}$ , we approximate the integral

$$n_{\rm X}^{\rm br} = \frac{1}{4\pi^2} \int_{|q_{\parallel}| < q_{\rm max}} N_{\rm 1s,q} \, {\rm d}^2 q \approx N_{\rm 1s,q=0} \frac{q_{\rm max}^2}{4\pi},\tag{13}$$

where the maximum value for the parallel component of the wave vector,  $q_{\max}$ , depends on the experimental set up, the spot size, etc. In the present paper, in accordance with Ref. 17, we count all excitons with  $q_{\max} < \frac{E_g}{\hbar c_0}$ , where  $E_g$  is the gap energy and  $c_0$  the vacuum velocity of light. Thus, all excitons emitting photons which can leave the barrier are counted as bright excitons. Alternatively, we could have used  $q_{\max} < \frac{E_g n}{\hbar c_0}$ , corresponding to all excitons which can emit photons into the substrate with refractive index n. However, since all photons with  $\frac{E_g}{\hbar c_0} < |q_{\parallel}| < \frac{E_g n}{\hbar c_0}$  experience total internal reflection at the substrate-air boundary, they can easily be reabsorbed by the quantum well and do not necessarily contribute to a decrease in exciton density.

The calculated fraction of bright excitons  $n_{\rm X}^{\rm br}/n_{\rm eh}$  necessary to reproduce the experimental  $\beta$  values is plotted in Fig. 4 (d). The temporal evolution clearly demonstrates that — for our material system and excitation conditions — excitons need several hundreds of ps to form and subsequently to relax down to the optically active  $q \approx 0$  state, where q is the exciton center-of-mass momentum. This time span is in full agreement with recent THz results and theoretical predictions<sup>4,13</sup>. We want to emphasize that the fraction of bright excitons is displayed. While this fraction monotonuously increases over 2 ns, the bright exciton density  $n_{\rm X}^{\rm br}$  itself exhibits a maximum around t = 800 ps. As an additional caveat, we also note that the value of  $\beta$  is extremely sensitive to the temperature deduced from the continuum PL. Therefore, the theoretically determined bright exciton numbers have been extracted carefully and are



FIG. 5: Same as Fig. 4 but for an initial carrier density of  $n_{\rm eh} = 7.9 \times 10^8 \, {\rm cm}^{-2}$ .

assumed to be correct, but due to temperature and other parameter uncertainties, the exact values could possibly be off by at most a factor of two.

Figure 5 displays equivalent data as Fig. 4, but now for a much lower initial carrier density of  $n_{\rm eh} = 7.9 \times 10^8 \, {\rm cm}^{-2}$ . While the qualitative behavior is very similar, a few differences must be pointed out. First of all, the temperature extracted from the continuum PL drops faster and approaches lower values than in the previous case of higher density. Secondly, the density falls off more slowly. While in Fig. 4 it drops by roughly a factor of 10 within 2 ns, it now drops by less than a factor of 7. As far as  $\beta$  is concerned, the pure plasma value is smaller by an order of magnitude compared to Fig. 4. Nevertheless, fewer excitons are needed to correctly fit the experimental data. Not only is the absolute value of  $N_{1s,q=0}$  smaller, but even the fraction of bright excitons shown in Fig. 5 (d) is smaller and displays a qualitatively different behavior than in Fig. 4. At later times, the excitons appear to decay quicker than the carrier density such that the ratio  $n_{\rm br}^{\rm br}/n_{\rm eh}$  exhibits a maximum after 600 ps.

### IV. CONCLUSIONS

Emission from the 1s-exciton resonance has been studied by means of time-dependent PL measurements. A microscopic theory has been applied to quantify the amount of bright 1s-excitons. The regime of very low, liquid-helium lattice temperatures shows the presence of an incoherent population of excitons, which dominates the 1s-emission dynamics in a wide range of carrier densities. A time of 600 to 800 ps is found necessary for exciton formation and relaxation to the lowest momentum states. With increasing lattice temperature a smooth transition occurs at about 30 K to a regime where the 1s-emission is completely due to the population of unbound electron-hole plasma states.

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