Luminescence patterns in photoexcited quantum wells: diffusion of the Coulomb plasma versus exciton superfluidity

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Abstract.

Inspired by experimental observations of ring-like luminescence patterns outside the laser spot in double [1] and single [2] quantum wells (QWs), we performed molecular dynamics (MD) simulations of the electron-hole plasma. Electron and hole density profiles generally follow the hydrodynamic two-fluid kinetic equations with non-trivial power laws for hole and electron velocities. While the MD or its kinetic approximation explain the inner ring formation, they fail to account for the macroscopic outer luminesce rings observed both in double and single QWs. Solving the Gross-Pitaevskii equation we show that in contrast to previous theoretical models describing the outer ring formation via conventional (normal state) diffusion, this phenomenon is a sequence of superfluidity of excitons in a dark state.

To describe the dynamics of hot electrons and holes, we use classical equations of motion [3]

\[
\begin{align*}
m^*_e \ddot{r}_e^i + \gamma_e \dot{r}_e^i &= \sum_{j \neq i} \frac{e^2 (r_e^i - r_e^j)}{|r_e^i - r_e^j|^3} - \sum_{k} \frac{e^2 (r_e^i - r_h^k)}{[(r_e^i - r_h^k)^2 + d^2]^{3/2}} + \xi_e, \\
m^*_h \ddot{r}_h^i + \gamma_h \dot{r}_h^i &= \sum_{j \neq i} \frac{e^2 (r_h^i - r_h^j)}{|r_h^i - r_h^j|^3} - \sum_{k} \frac{e^2 (r_h^i - r_e^k)}{[(r_h^i - r_e^k)^2 + d^2]^{3/2}} + \xi_h,
\end{align*}
\]

where vectors \( r_e^i \) and \( r_h^j \) are in-plane positions of \( i \)-th electron and \( j \)-th hole, \( e \) is the electron charge, and \( d \) is the interlayer distance. The left-hand side of Eqs.(1) accounts for inertia of electrons and holes with their effective masses \( m^*_e \) and \( m^*_h \) respectively as well as for the momentum damping (described by constants \( \gamma_e \) and \( \gamma_h \)) due to interaction with acoustic phonons. The right-hand side accounts for the in-layer electron-electron and hole-hole Coulomb repulsions, the inter-layer electron-hole attraction, and random forces, \( \xi_e,h \) describing electron and hole diffusion. Here \( \langle \xi_e \rangle = \langle \xi_h \rangle = 0 \) and \( \langle \xi_e(0) \xi_e(t) \rangle = \langle \xi_h(0) \xi_h(t) \rangle = \delta(t) \). In simulations we normalized time by \( t_s = m^*_e \mu_e / e \) and all distances by \( r_s = \sqrt{m^*_e \mu_e} \) with electron mobility \( \mu_e \).

The optical excitation of electrons and holes was modeled by generating the particles in random positions inside the excitation spot (i.e., within interval \(-x_0 < x < x_0\)) with a rate \( p \). Initial velocities of electrons were also chosen randomly. An exciton formation happened if an electron and a hole were close enough to each other, \( |r_e - r_h| < a \), where \( a(d) \) is a phenomenological
Figure 1. (a): Qualitative comparison of distributions of the electron and hole velocities and densities predicted by the hydrodynamical model (top panel) with the simulated distributions (the distance from the spot $x$, densities, $n_e$, $n_h$, and velocities, $v_e$, $v_h$, as well as density of exciton formation events, $n_{lum}$, are given in arbitrary units). (b): The exciton superfluid momentum as a function of the distance (arb. units) from the laser spot obtained using Eq.(3) for three different values of the decay rate $\beta = \gamma/2\mu = 10^{-2}$, $10^{-3}$, $10^{-4}$ in 1D and 2D geometries [4].

Some of our 1D results obtained by simulating the above classical equations of motion without the random forces ($\xi_{e,h} = 0$) are shown in Fig.1a. As soon as we begin simulations switching on the laser pumping, the number $N$ of excited electrons and holes starts to increase and finally saturates after a short time. We plot the spatial distributions of electrons, holes and exciton formation events by averaging the number of particles (events) within a certain short spatial interval. One can see in Fig.1a that electrons and holes are spatially separated: holes are accumulated close to the excitation spot while electrons are mostly located far away from the spot. In the region of the overlap of electron and hole densities the luminescence peak occurs at a certain macroscopic distance from the spot. Qualitatively this picture is somewhat similar to the earlier explanations of luminescence patterns within purely diffusion model, but here we did not make any ad-hoc assumptions (e.g. assuming an electron flux from external parts of the sample, and/or neglecting the Coulomb forces). As seen from Fig. 1a, the electron average velocity decreases with $x$ monotonically while the hole velocity surprisingly increases with $x$.

As shown in Fig.1a most of our MD simulation results can be reproduced in a simplified hydrodynamic model derived by truncating the Bogoliubov - Born - Green - Kirkwood - Yvon
(BBGKY) hierarchy of many-particle distribution functions to one-particle distributions,

\[ J^2 \frac{dn_e^{-1}}{dx} + \gamma_e J = \frac{n_e F_e^C}{m_e^*} \]
\[ J^2 \frac{dn_h^{-1}}{dx} + \gamma_h J = \frac{n_h F_h^C}{m_h^*}. \]  

(2)

with a constant \( J \), which is the current for both electrons and holes, and the mean-field Coulomb forces, \( F_{e,h}^C \).

We have also studied how electron, hole and luminescence distributions depend on the laser (pump) intensity by changing the e-h generation rate \( p \). It turns out that the position of the luminesce peak depends very weakly on \( p \): slightly increasing at low \( p \) and then starting to decrease. Thus, our normal state MD of the electron-hole plasma fails to explain the experimental observations [1, 2] of a strong dependence of the outer (external) ring radius on the laser intensity, but it is compatible with the properties of the inner ring observed in double QWs [1]. So we can conclude that the ring observed in our simulations can explain the inner ring where excitons with a finite momentum are formed.

To explain the external macroscopic rings with the radius of about a few hundreds microns [1, 2] we propose [4] that excitons condense in a coherent superfluid with a finite momentum from electron-hole plasma at the inner ring since the electron-hole plasma expands inside the spot due to the Coulomb repulsions. If the lattice temperature is below the Berezinskii-Kosterlitz-Thouless (BKT) transition temperature of 2D excitons, this new state of matter allows excitons to travel in a dark state on a macroscopic distance from the laser spot producing photoluminesces (PL) rings far away from the photo-excited region as observed in Refs.[1, 2]. To describe a free superflow of the 2D Bose-liquid of decaying excitons we apply a generalized Gross-Pitaevskii (GP) equation [5] for the order-parameter, \( \psi(R) \) taking into account the exciton recombination rate \( \gamma/\hbar \), as originally proposed by Keldysh [6]:

\[- \left( \frac{\hbar^2}{2m_{ex}} \Delta + \mu \right) \psi(R) + V|\psi(R)|^2 \psi(R) - i\frac{\gamma}{2} \psi(R) = 0. \]

(3)

Here \( \mu \) and \( V \) conveniently parameterize the average superfluid density and the short-range repulsion, respectively. To determine location of luminescence rings, we use that excitons can radiate in 1D and 2D by resonant emission of photons only if their momentum is inside the photon cone, \( K < K_c \). The solutions of the GP equation are shown in Fig.1b for 1D and 2D geometries.

In dimensional units the PL external ring radius is found as \( R_{ring} \approx \sqrt{\hbar v \tau_{ex} (16\pi d \alpha n/m_{ex})^{1/2}/\gamma} \).

Importantly \( \gamma \) in this estimate is the nonradiative recombination rate since the coherent excitons have their momentum outside the photon cone, \( d \) is the screening radius in the electron-hole plasma, \( \tau_{ex} \) is the exciton formation time, \( v \) is the characteristic speed of photocarrires, \( m_{ex} \) is the exciton mass and \( n \) is the photocarrier density. Remarkably, using the realistic material parameters yields \( R_{ring} \) about 300 \( \mu m \) for the photoelectron density \( n = 10^{10} \) cm\(^{-2} \), explaining the external PL rings [4].

References
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