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Dynamics of the inner ring in photoluminescence of GaAs/AlGaAs indirect excitons

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Abstract.

A theoretical description of the diffusion, thermalization and photoluminescence of indirect excitons in low temperature (≈ 1K) GaAs/AlGaAs coupled quantum wells is compared with experiments on their photoluminescence dynamics. The results shown in this contribution demonstrate a highly accurate agreement between the two. We concentrate on two key features seen in the photoluminescence pattern: the formation of an inner ring around a tightly focused laser excitation spot and a rapid increase in the intensity from the excitation spot immediately after laser termination - the PL-jump. These striking effects are explained in terms of the diffusion and relaxation thermodynamics of indirect excitons.

1. Introduction

In coupled quantum wells (CQWs), two types of exciton may be created. Firstly, the direct type where the electron and hole are in the same quantum well (QW), and secondly the indirect type where the electron and hole reside in adjacent QWs. When a voltage is applied across the QWs, the indirect exciton population dominates over that of the direct type as the latter is less energetically favourable and decays more rapidly. Electrons and holes tunnel into their respective wells and relax into the indirect exciton state. Such a system is particularly useful for studying the transport of a statistically-degenerate Bose gas in solids. This is because, (i) the spatial separation of the electron and hole wavefunctions reduces their overlap and increases their lifetime by orders of magnitude, and (ii) excitons in CQWs cool very efficiently as they interact with a bath of bulk longitudinal-acoustic (LA) phonons at the lattice temperature, \( T_b \). Consequently, indirect excitons travel large distances from their point of creation and cool close to their degeneracy temperature, \( T_0 \). For indirect excitons, which from here we consider only, \( T_0 = (2\pi\hbar^2 n_x)/(gM_x k_B) \approx 1K \) for 2D concentration \( n_x \approx 10^{10}\text{cm}^{-2} \), spin degeneracy factor \( g = 4 \) and exciton mass \( M_x = 0.22m_0 \) (\( m_0 \) is the free electron mass).

In recent years, a series of experiments has uncovered a number of fascinating features in the exciton kinetics and emission patterns. These include an internal ring - a peak in the intensity that forms around a tightly focused laser excitation spot [1, 2], and the PL-jump - a large increase in the intensity emitted from the excitation spot immediately after termination of the laser pulse [3]. Other features observed are localized bright spots [1], an external ring [1, 2, 4, 5] and a macroscopically ordered state [1]. Further details of these can be found in Ref.[6].
A recent report [7], claimed that the features observed in this type of experiment are the result of ambipolar diffusion of an unbound electron-hole (e-h) plasma rather than exciton transport and cooling. This significantly different explanation of the inner ring is invalid for our current work where the exciton concentration $n_x$ completely dominates over that of free e-h pairs. We justify this using the quantum mass action law (QMAL). For effective exciton temperatures ($T \leq 6K$) relevant to the experiments, the QMAL gives the free e-h concentration $n_{e,h} \approx \left( \frac{m_e m_h k_B T}{\hbar^2} \right) \exp[-\epsilon_x/(2k_B T)] \ll n_x$ ($\epsilon_x$ is the exciton binding energy and $n_{e(h)}$ is the electron(hole) mass). This view is consistent with experiments where the observed PL linewidth from CQWs corresponds to exciton decay rather than e-h annihilation. For more details, see Refs. [8, 9]. Further, the mean-field energy is discussed against the correlation energy in [8]. It is shown that for $n_x \geq 10^9 \text{cm}^{-2}$, the former dominates over the latter and therefore only the mean-field energy is used in our model. Hence the exciton interpretation is the correct one to model our experimental data.

We present here time resolved measurements of the inner ring in the PL pattern from experiments [9]. A CQW structure consisting of two $8 \text{nm}$ GaAs layers separated by a $4 \text{nm}$ Al$_{0.33}$Ga$_{0.67}$As barrier was illuminated by a focused laser pulse of duration $500 \text{ns}$. More details of the experimental setup can be found in [9]. We show that the temporal evolution of the inner ring is predicted by a set of non-linear differential equations that govern their diffusion, thermalization and PL dynamics. Further, the model accurately reproduces the dynamics of the PL-jump for which we also show experimental data. The outer ring and localized bright spots are not considered here since they have no effect on the kinetics of the inner ring.

2. Theory and results

The transport of excitons is governed by the following non-linear diffusion equation for $n_x$ [10]:

$$\frac{\partial n_x}{\partial t} = \nabla \cdot [D_x \nabla n_x + \mu_x n_x \nabla (u_0 n_x)] - \Gamma_{opt} n_x + \Lambda,$$  \hspace{1cm} (1)

Here, $D_x(n_x, T)$ is the diffusion coefficient, $u_0 n_x$ is the exciton dipole-dipole interaction energy (the exciton line blue shift in the PL spectrum) and $\mu_x$ is the exciton mobility which is related to the diffusion coefficient through the generalized Einstein relation [10], $\mu_x = D_x (e^{T_0/T} - 1)/(k_B T_0)$. $\Lambda(r, t)$ is the generation rate of excitons from the binding of photoexcited e-h pairs as described by the QMAL. Here, $r$ is the distance from the center of the excitation spot. $\Gamma_{opt}(n_x, T)$ is the exciton optical decay rate [11]. This is calculated according to the fact that only the lowest energy (bright) exciton states inside the photon cone may decay to emit light. This means initially hot excitons must cool before decaying radiatively.

The diffusion equation (1) was solved with a time-space varying temperature according to

$$\frac{\partial T}{\partial t} = S_{\text{phonon}} + S_{\text{pump}} + S_{\text{opt}}.$$  \hspace{1cm} (2)

The term $S_{\text{pump}}(n_x, T, \Lambda)$ accounts for the initially hot, injected excitons that cause a net heating of the system. As excitons transport through the lattice, they are cooled by interactions with a bath of bulk LA phonons. This effect is evaluated by $S_{\text{phonon}}(n_x, T)$. In the absence of heating, i.e., away from the excitation spot or after laser termination, this mechanism rapidly brings excitons into thermal equilibrium with the lattice. Since only the lowest energy excitons may decay optically, they are removed by PL emission causing heating of the system. This is accounted for by $S_{\text{opt}}(n_x, T)$. The terms $S_{\text{pump}}$, $S_{\text{phonon}}$ and $S_{\text{opt}}$ are detailed in Refs. [9, 12].

In-plane diffusion of excitons can be suppressed by QW disorder which arises from imperfections and alloy fluctuations in the GaAs/AlGaAs samples used. For $n_x \geq 10^9 \text{cm}^{-2}$, the accumulation of excitons strongly screens the disorder (see Ref. [10]). This is included using
Figure 1. Exciton kinetics after activation of the laser pulse. (a)-(e) show the calculated spatial profiles at various times for the density, diffusion coefficient, temperature, optical decay rate and PL intensity. $t = 0$ corresponds to laser activation. The laser profile, $\Lambda$, is the thin red dashed line in (e). Shown in (f) are measurements of the spatial profile of the PL intensity from experiments. All parameters used in the model are chosen to match experimental conditions so that a quantitative comparison can be made. Both experimental and calculated PL intensities from the excitation spot and the ring position as a function of time are shown in (g).

a thermionic model for the diffusion coefficient, $D_x = D_x^0 \exp[-U_0/(k_B T + u_0 n_x)]$, where $U_0$ is the QW disorder potential and $D_x = D_x^0$ in the absence of disorder.

Equations (1) and (2) with the non-linear diffusion coefficient and decay rate $\Gamma_{opt}(n_x, T)$ describe the evolution of the creation, transport, thermalization and optical decay of excitons in CQWs. An explicit finite difference scheme was used to integrate the differential equations in the space-time $(r, t)$ domain. In Fig 1 (a), numerical solutions of Eq.(1) subject to optical generation of excitons are shown. It can be seen that within 30 ns, excitons diffuse tens of micrometers away from the excitation spot. Such rapid diffusion occurs because for high densities, QW disorder is strongly screened by exciton-exciton interaction leading to a larger diffusion coefficient, see Fig 1 (b). The sharp contrast in density for large values of $r$ is a consequence of the reduction in diffusion coefficient as QW disorder is not as effectively screened for lower densities.

Temperature profiles of the system are given in Fig 1 (c). Inside the excitation spot, the profile closely follows that of the generation rate, $\Lambda$ (thin dotted line in Fig 1 (e)). Outside, excitons are in thermal equilibrium with the lattice which is at the helium bath temperature, $T_b = 1.4\, K$. The effect of temperature on the exciton decay rate, $\Gamma_{opt}$ is seen in Fig 1 (d). At the excitation spot, heating due to the laser suppresses decay and hence the PL intensity is reduced. The result is the formation of a bright ring in the PL pattern around the excitation spot as shown in Fig 1 (e).

Data obtained from experiment is presented in Fig 1 (f). The PL intensity emitted from GaAs/AlGaAs CQWs was collected with 4 ns time resolution. (All basic parameters in the theoretical model were chosen to match experimental conditions and only a few control parameters were used to give a best fit between experimental and theoretical data. For more details, see Ref. [9].) An accurate agreement between Fig 1 (e) and (f) is clearly seen. There is also a strong agreement in the theory/experimental PL dynamics, plotted together in Fig 1 (g).
Figure 2. Dynamics of the PL-jump. (a)-(d) show the calculated time dependence of the temperature, decay rate, density and PL emission of excitons at the excitation spot and ring position after termination of the laser. Laser termination occurs at \( t = 500 \text{ ns} \) (shown by the vertical blue dashed line). The lines connected by crosses/triangles in (d) are the PL intensity measured in experiment for comparison.

Fig 2 shows exciton kinetics after laser termination. Without heating by the laser \( (S_{\text{pump}} = 0) \), the population is allowed to thermalize with the lattice. Although the system is heated by exciton decay, its effect is relatively weak compared to the highly efficient cooling by phonon interactions. Within 2 ns, the temperature at the excitation spot drops close to the lattice temperature, see Fig 2 (a). Consequently, the decay rate, \( \Gamma_{\text{opt}} \) shown in Fig 2 (b), increases considerably and the large population of excitons at the excitation spot begins a fast optical decay, see Fig 2 (c). As shown in Fig 3 (d), this sudden increase in PL intensity, the PL-jump, is observed in experiments and predicted by the model. The intensity approximately doubles in magnitude before beginning exponential decay. The discrepancy between measured and theoretical results for the magnitude of the PL-jump is due to the limited temporal resolution of the imaging device.

3. Conclusions
The theoretical model of exciton dynamics accurately predicts the formation of the inner ring observed in experiments. As excitons screen QW disorder, they quickly diffuse away from the excitation spot where rapid cooling leads to a large increase in their decay rate and hence PL intensity. Termination of the laser removes the main heating mechanism causing the high population at the excitation spot to thermalize with the lattice. This results in the PL-jump, also observed experimentally. The experimental and theoretical results are in quantitative agreement.

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