Available online at www.sciencedirect.com
SCIENCE DIRECT®
Chemical Physics 318 (2005) 104–117
Chemical Physics
www.elsevier.com/locate/chemphys

Excitonic effects in the luminescence of quantum wells

B. Deveaud [a,*], L. Kappei [a], J. Berney [a], F. Morier-Genoud [a], M.T. Portella-Oberli [a], J. Szczytko [a], C. Piermarocchi [b]

[a] Institut de Photonique et Electronique Quantiques, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH1015 Lausanne, Switzerland
[b] Department of Physics and Astronomy, Michigan State University, East Lansing, MI, USA

Received 3 February 2005; accepted 13 June 2005
Available online 19 August 2005

Abstract

We report on the origin of the excitonic luminescence in quantum wells. This study is carried out by time-resolved photoluminescence experiments performed on a very high-quality InGaAs quantum well sample in which the photoluminescence contributions at the energy of the exciton and at the band edge can be clearly separated and traced over a broad range of times and densities. This allows us to compare the two conflicting theoretical approaches to the question of the origin of the excitonic luminescence in quantum wells: the model of the exciton population and the model of the Coulomb correlated plasma. We measure the exciton formation time and we show the fast exciton formation and its dependence with carrier density. We are also able to give the boundaries of the Mott transition in our system, and to show the absence of observable renormalization of the gap below the onset of this transition. We detail the characteristics of the trion formation and evidence the possible formation of both positive and negative trions in the absence of any resident free carrier populations.

Keywords: Excitonic effects; Luminescence; Quantum wells; Trions; Renormalization; Mott transition

1. Introduction

Excitons in quantum wells have been a very active subject of research, and Daniel Chemla has contributed in a major way to the success story of these quasi-particles that everybody would think will stay forever confined in the laboratories of Physics Departments of Universities, just because these are unstable at room temperature. However, even if the lifetime of excitons at room temperature in quantum wells is as short as 300 fs, they nevertheless show clear resonance features in absorption allowing a direct observation of the resonance and the possible use of excitons in real devices [1]. Such resonances may be used in a number of photonic devices such as for example the SEED device, which was proposed by Miller and Chemla [2], and is based on an effect of major importance in quantum wells, the quantum confined Stark effect [3]. The group of Daniel Chemla also introduced, shortly later, the idea of the influence of a virtual exciton population on the resonance properties of the excitons [4,5], the so-called Optical Stark effect.

Excitons may also be observed in luminescence, and Weisbuch and co-workers [6] were the first to introduce the idea that, contrary to bulk semiconductors, luminescence properties of quantum wells were dominated by free exciton luminescence. This corresponds to a major difference from the case of excitons in bulk semiconductors where polaritonic effects do prevent the direct recombination of excitons [7] each exciton being strongly coupled with one and only one photon mode. In quantum wells, the breakdown of the translational symmetry in the growth direction allows each exciton

* Corresponding author. Tel.: +41216935496; fax: +41216934525.
E-mail address: benoit.deveaud-pledran@epfl.ch (B. Deveaud).

0301-0104/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.chemphys.2005.06.045

to be coupled to a continuum of photons, thereby allowing the radiative decay of the excitons within the radiative zone defined by the conservation of the in-plane wavevector between the exciton and the photon. The radiative decay of these excitons is very fast, as first proposed theoretically by Agranovich and Duboskii [8] and checked experimentally much later on [9].

The absence of any clear observation of free carrier luminescence in the spectra of quantum wells, except for the case of very high excitation density, and the very fast build-up of the exciton line after non resonant excitation [10] has led Stefan Koch and co-workers [11] to propose that luminescence at the exciton energy might not be the signature of an exciton population. Indeed, the polarization linked with the presence of an uncorrelated electron–hole plasma may lead, in particular at high enough densities, or at short enough times, where the strict energy and momentum conservation rules are relaxed, to the emission of light at the energy of the resonance of the system, i.e., the energy of the excitons. Here, we will review our recent attempts, correlated with the work performed in the group of Daniel Chemla by Kaindl et al. [12], to understand whether or not a real population of excitons was indeed present in quantum wells, and what would be the relation between such a population and the actual luminescence properties of the quantum well. The paper of Kaindl, Chemla corresponds to a very interesting idea that is to probe the exciton population through the time resolved measurements of the tera-Herz absorption of excitons. The technique has the major advantage to be sensitive to the whole population of excitons. The drawback of this difficult technique is the narrow density range where it can be applied, and the poor sensitivity to the free carrier population. Our results are based on a series of detailed experiments with a set of samples of unprecedented quality, allowing to observe clearly the free carrier transitions as a separate peak. Through the decay of this population that we can follow very accurately, and knowing that, in such samples, the exciton formation is the dominant decay mechanism we can directly infer the exciton population even if the luminescence intensity at the exciton energy does not reflect in any direct way the exciton population. Our measurements are therefore quite complementary to those of Kaindl and Chemla and, through the use of a simple rate equation analysis taking into account the main ingredients of the physics of the system, we get numbers for the exciton formation in reasonable agreement with those of Kaindl et al. over the density range where they succeed in performing experiments. We also study, on the same set of samples and making use of their particular quality, the Mott transition as well as the dynamics of trion formation. We show that the Mott transition is a smooth crossover, occurring for densities ranging from 10^{10} to 10^{11} cm^{-2}. We demonstrate that the trion formation results from both bimolecular and trimolecular binding processes.

2. Experiment and samples

We have selected a particular sample, because of its unusually high quality, although similar samples were obtained from the same machine, or from other machines giving equivalent results [13]. The sample used for this study is a single $In_xGa_{1-x}As$ 80 Å quantum well (QW), with an indium content of about $x = 5\%$ grown by molecular-beam epitaxy. This QW is embedded in the middle of a GaAs layer of total mean thickness λ (where λ corresponds to the wavelength of the excitonic resonance in the QW) grown over a 10 period distributed Bragg reflector (DBR). This DBR allows first to directly measure the absorption of the sample in the reflection configuration without any preparation of the sample. It also increases the optical coupling of the QW, but does not disturb appreciably the shape of the observed PL spectrum, because the resonance mode has a spectral width of about 40 nm, which is at least one order of magnitude larger than the width of any of the structures observed in luminescence. Such a DBR slightly changes the radiative properties of free carriers, but does not affect their relaxation properties, which we are studying here.

The sample is cooled down to a temperature of 4.5 K in an helium bath cryostat. The position of the focal point of the optical excitation is controlled with precision of about 1 μm. In cw mode, the sample is excited by a cw Ti:Sapphire laser. For time-resolved luminescence spectroscopy, we use spectrally filtered 100 fs pulses from the same Ti:Sapphire laser working in the mode-locked regime. The interference spectral filter used as a pulse-shaper allows a spectral resolution of about 0.8–1.0 nm, and a temporal duration of about 1.2 ps. We used a 50 cm monochromator with a 600 grooves per mm grating. The spectrum is recorded with a charge-coupled device (CCD) camera in the cw mode and with a streak camera in the time-resolved experiment (resolution of 3 ps, photon-counting mode). The temporal resolution of the whole setup is limited to about 10–20 ps, due to the dispersion of the grating, allowing a 0.1 meV spectral resolution.

What is particularly appealing in that particular sample is the fact (see Fig. 1) that we clearly observe in the cw spectrum transitions corresponding to the 1s exciton, the 2s exciton the free electron–hole pairs and also the trions (not clearly resolved in Fig. 1, due to the specific excitation energy) [14]. Proper identification of the structures observed in our absorption and photoluminescence experiments is very crucial for our reasoning. As our assignments differ from the identification proposed