Optical spectroscopy of quantum wells under high excitation

Autor(en):   **Bongiovanni, G. / Moser, J. / Staehli, J.L.**

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OPTICAL SPECTROSCOPY OF QUANTUM WELLS UNDER HIGH EXCITATION

G. Bongiovanni, J. Moser and J.L. Staehli: Institut de Physique Appliquée, Ecole Polytechnique Fédérale, PH-Ecublens, CH-1015 Lausanne, Switzerland.

Abstract: We briefly survey the properties of the optically excited electron-hole fluid confined in a quantum well. It is easy to substantially populate the different subbands, and on the low energy transitions optical gain develops in a wide spectral region. The observed particle distribution often deviates from that predicted by Fermi statistics. We discuss the many-body interactions causing gap shrinkages and governing the establishment of thermal quasi equilibria among the carriers.

INTRODUCTION.

The opportunity to confine electrons in quantum wells (QW) opened the possibilty to study electron-electron (e-e), electron-hole (e-h) and electron-phonon (e-ph) interactions in systems approaching the two dimensional (2d) limit. The results of our luminescence and transmission measurements on highly excited QW show indeed that all these interactions have to be accounted for to fully describe the properties of the confined e-h fluid.

EXPERIMENTAL.

The experiments were performed in an undoped multiple QW structure of GaAs/-(GaAl)As grown by molecular beam epitaxy. The wells have a width of 122 Åand the structure contains only 29 wells which ensures a uniform excitation in depth. The sample was immersed in superfluid He, and the e-h pairs were excited by a frequency doubled and pulsed YAG:Nd laser (pulse duration about 50 ns). The energy of the laser photons was 2.33 eV which is higher than the gap in the (GaAl)As barriers. A dye jet, pumped by the same YAG:Nd laser, served as a broad-band light source for the transmission measurements. Lock-in and boxcar techniques were employed to measure transmission and luminescence spectra contemporaneously.

In figure 1 absorption and luminescence spectra are shown for different excitation in-

tensities. At low pump levels, sharp exciton peaks are visible at the subband edges. As the excitation intensity is increased, the changes observed on the transitions between the lowest subbands (labelled 11 in figure 1) are different from those between higher subbands.
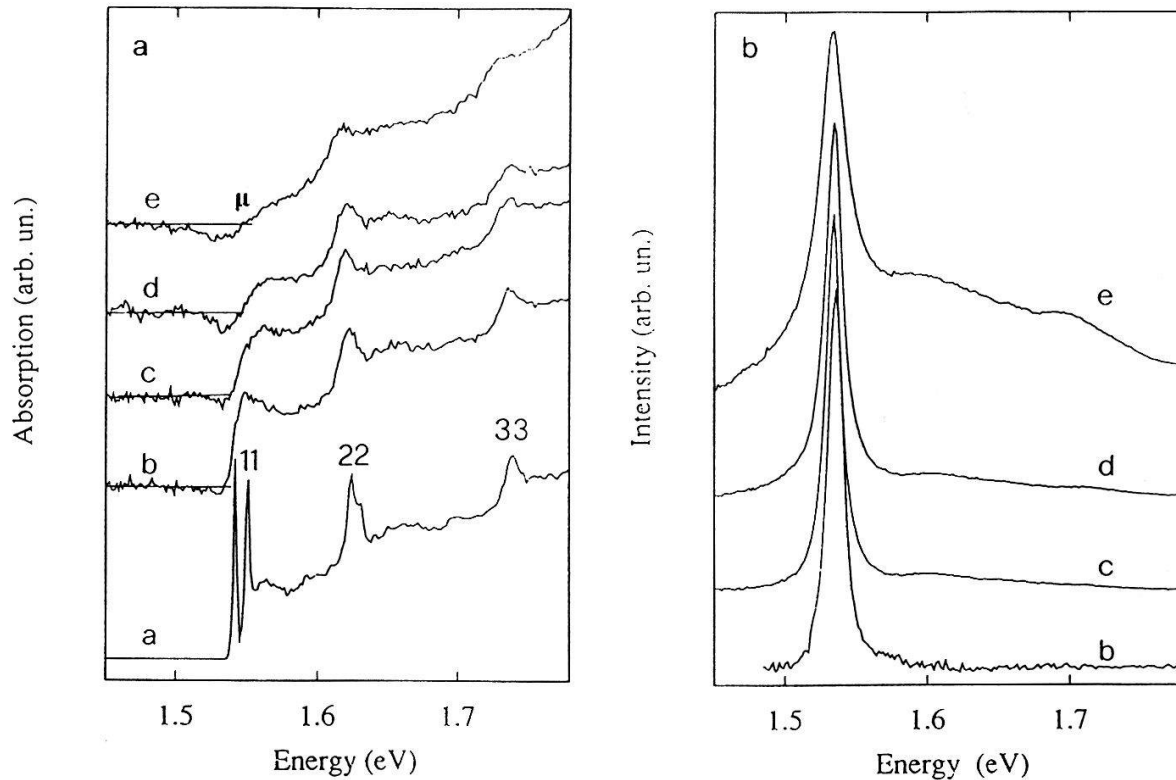
Figure:1. Absorption (a) and luminescence (b) spectra of a $GaAs/Al_{1-x}Ga_xAs$ multiple QW structure at different excitation levels. The lattice temperature is 2K. The excitation intensities are as follows; a: 0; b: 4 ; c: 15; d: 80; and e: 400 $kWcm^{-2}$. 11, 22 and 33 label the the electron and hole subbands between which the transitions take place. μ indicates the spectral position of the chemical potential of the 11 pairs (supposing thermodynamic equilibrium).

a) 11 transitions. In the absorption spectra, the narrow exciton peaks quickly disappear, even if the e-h correlation which enhances the transition probability mainly near the absorption edge remains large (see eg the spectrum with $I_{EX} = 4$ kWcm^{-2}) [1]. At higher excitations also the transitions between continuum states are partially bleached, the enhancement effect weakens, and at lower energies the spectral region with optical gain widens. In the luminescence spectra, at the wavelengths where gain is observed, a strong peak appears.

b) Transitions between higher subbands. In the absorption spectra, relatively strong excitonic peaks are present up to almost the highest excitations, and they are shifted to

the red by a few meV. The absorption by the continuum transitions is reduced while the intensity of the luminescence emission increases.

SUBBAND RENORMALIZATION.

The experimental results can be interpreted as follows. At weak excitation almost all e-h pairs relax toward the fundamental 11 band edge. Exchange and screening effects reduce the binding energy of the 11 exitons, and at the Mott concentration (which is different for heavy and light hole excitons), they merge into the continuum [2]. However in 2d screening alone is not sufficient to drive the exciton binding energy to zero. In other words, exchange effects of dissociated kk pairs are essential to reduce the continuum edge below the kk bound states. This explains why the 11 excitons disappear while the 22 and 33 excitons persist up to quite high excitations, as long as the higher subbands are only weakly populated. Inter-subband exchange is negligible [3]. The disappearance of the 11 exciton at lower pump intensties has also been explained by phase space filling [4].

Once the density of the 11 pairs is high enough population inversion occurs and optical gain is observed. In a system in thermodynamic equilibrium the crossover from gain to absorption takes place at $\hbar\omega = \mu$, as shown in figure 1a. The strong peak in the luminescence spectra is found in the same spectral region where the gain is observed. Therefore we believe that this peak is caused by stimulated emission travelling in the well plane, and that crystal imperfections scatter it out of the sample.

The strongest gain occurs at energies below the 11 heavy exciton, evidencing the band gap reduction caused by exchange and correlation interactions. Further, the absence of sharp edges in the experimental spectra shows that the single particle states are also collision broadened [5]. Thus, to extract reliable values of the gap reduction from the optical spectra an elaborate analysis must be performed [6]. The results show that the band gap shrinkage depends on the well width and, if it is measured in exitonic units, is smaller than in 3d [7].

The renormalization of the higher subbands can roughly be estimated from the red shift of the corresponding exciton peaks as long as their oscillator strengths (and consequently their binding energies) vary only slowly with pump intensity [8,9]. For the absorption spectrum d the red shifts are 4 and 2 meV for the 22 and 33 subbands, respectively. The gap reduction between the lowest 11 subbands instead is much larger, it can be estimated to about 17 meV. This experimental result rules out the hypothesis of a rigid shift of all the subbands together [10]; again this is due to the lower population of the higher subbands

and due to the fact that inter-subband exchange is negligible. However, a quantitative comparison between experiment and theory is still lacking.

CARRIER DISTRIBUTION.

In thermodynamic equilibrium characterized by a temperature T_{eh} and a chemical potential μ, the ratio $R(\hbar\omega)$ between absorption coefficient $\alpha(\hbar\omega)$ and spontaneous luminescence intensity $I_L(\hbar\omega)$ is proportional to the inverse Bose function $f_B^{-1}(\hbar\omega)$; this ratio does not directly dependend on the joint density of states [5]. Thus, deviations from thermodynamic equilibrium can be monitored easily and quantitatively. As is visible in figure 2, the natural logarithm of the experimentally determined ratio $R(\hbar\omega)$ shows more structure than one would expect at thermal equilibrium. In particular $lnR(\hbar\omega)$ shows steps and changes of the slope at the onsets of the transitions between higher subbands.
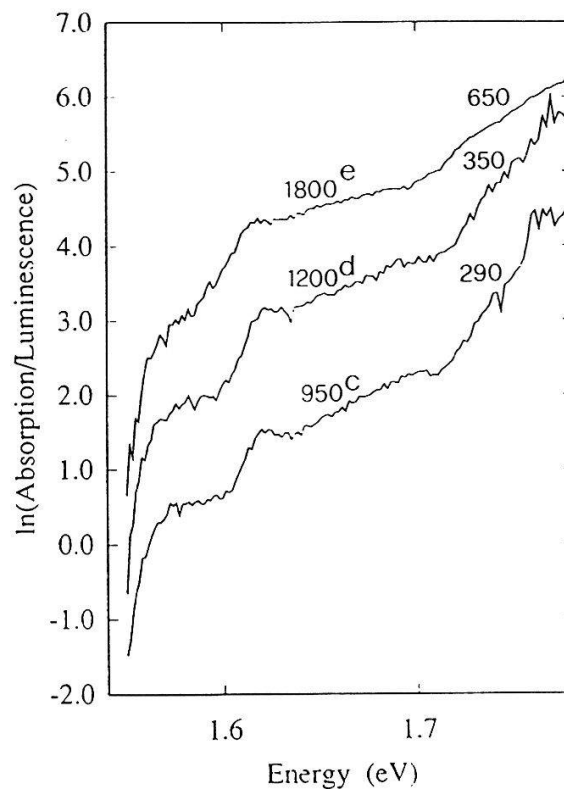
Figure:2. Natural logarithm of the ratio $R(\hbar\omega)$, absorption coefficient $\alpha(\hbar\omega)$ to luminescence intensity $I_L(\hbar\omega)$. The numbers on the curves are estimates of the carrier "temperatures" (in K) of the different subbands, they were determined by just reading the slope of $R(\hbar\omega)$. The letters have the same meaning as in figure 1.

In the experiment the e-h pairs are excited at an energy which is higher than the gap of the $Al_{1-x}Ga_xAs$ barriers. In the subsequent plasma thermalization the e-ph, e-h and e-e collision processes play a fundamendal role. The photon excess energy is partially transferred to the lattice via emission of longitudinal optical phonons. Since the collision time between particles in the same subband [11] is at least one order of magnitude shorter than that of e-ph scattering [12], an intra-subband quasi equilibrium is rapidly reached. However the inter-subband relaxation time is quite long [12], allowing the establishment of different equilibria in the different subbands. This seems to be confirmed by the experimental results in both the low and high excitation regimes, as shown in figure 2. The "temperatures" attributed to each subband are just rough estimates, better values may be obtained through a complex multi-band fit.

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