Coulomb Correlation and Band Gap Renormalization at High Carrier Densities in Quantum Wires

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We have studied the luminescence of narrow quantum wires at photoexcitation densities of up to $\sim 3 \times 10^6$ cm^{-1}. We show that even at these densities, which are well above the expected Mott density of 8×10^5 cm^{-1}, excitonic recombination dominates over other recombination channels in stark contrast with the behavior of quantum wells and bulk structures at equivalent densities. As we observe no significant shift in the peak energy with density, an upper limit to the band gap renormalization can be set. [S0031-9007(97)03044-5]

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Excitonic effects in a one dimensional (1D) system are expected to be even more significant than they are in two and three dimensional systems [1–3]. An enhancement of excitonic correlations [4] and an increased oscillator strength [5] have been observed experimentally. The question that then beckons is, up to what densities will excitons remain stable in quantum wires (QWRs); put otherwise one may wonder at what density the Mott transition occurs in optically excited QWRs. A fascinating aspect of high density studies, the Mott transition has been an active field of research over the past three decades [6], and there remain a number of open questions regarding this transition in three, two, and one dimensions [7]. In QWRs the transition is expected to occur when the insulating excitonic phase transforms into a conducting free electron-hole (*e-h*) plasma at a carrier density of about 8×10^5 cm^{-1} [3]. This transition has so far not been observed in optical studies, although it would be most interesting to probe it and associated effects such as a possible hysteresis in the Mott density [8].

Semiconductor QWRs have also been actively studied, as it is expected that the singularity in the density of states in a quasi-1D system may lower threshold current densities, thus improving the performance of semiconductor lasers [9–11]. The common procedure of computing the optical gain using a $1/\sqrt{E}$ density of states is, however, incomplete as a number of effects such as inhomogeneities, Coulomb interactions, and many body effects are neglected. A better understanding of the recombination of a dense *e-h* plasma in QWRs is thus called for, and the subject has attracted considerable attention of late [4,12–16].

Recent progress in the growth of semiconductor nanostructures has made available wires of good quality which should open the way for rigorous studies that probe the different interactions that occur in quasi-1D systems. It is then unfortunate that the picture that emerges from the published literature on high density phenomena in QWRs is confused and contradictory. A prime example of such ambiguity is the issue of band gap renormalization (BGR). Experimental reports range from evidence for a very large band gap energy shift [12,15], to no shift at all [4,13,14], with some authors observing shifts only due to the carriers in other subbands [16]. The theoretical results are equally ambiguous. Some models predict a very large BGR [17–19], even larger than in two dimensions (2D), while others emphasize the role of Coulomb correlations and excitons in 1D structures [1–3].

It is the purpose of this Letter to try to clarify the experimental situation and further our understanding of Coulomb correlations in 1D systems. In order to do this we studied quantum wires of the highest available quality and followed a careful experimental procedure that will be described below. Our results can be stated in a nutshell: We have evidence that *no measurable BGR is observed in QWRs, and most of the recombination is due to excitons, even at very high densities.*

The experiments were performed on V-groove GaAs quantum wire samples grown by low pressure organometallic chemical vapor deposition on patterned substrates [14]. That the samples studied are indeed of very high quality is established, notably by examining their photoluminescence (PL) and photoluminescence excitation (PLE) spectra, in different publications [20,21]. Their typical characteristics are (i) well resolved lateral subbands (typical electron subband spacing >20 meV), with clear polarization anisotropy due to valence band mixing, (ii) small inhomogeneous broadening ($\sim$8 meV), (iii) small Stokes shift ($\sim$6 meV), and (iv) very deep confining potential ($\sim$150 meV for electrons).

The samples were excited by 1–3 ps dye-laser pulses with an average power density of approximately 500 W cm^{-2} at 568 nm (this corresponds to a peak power of $\sim$2 MW cm^{-2}). At this wavelength we were exciting carriers in the wire and barrier. However, experiments performed at an excitation wavelength of 760 nm (thus only creating carriers in the wire) and a power density of 5 kW cm^{-2} yielded identical results. The time resolved PL spectra were acquired using a time correlated photon counting setup [22] with a resolution

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of about 50 ps. The luminescence was carefully imaged and spatially filtered, and consequently only the central part of the excited spot, in which the excitation density is homogeneous, was selected. The successive time delayed spectra recorded with this technique correspond to decreasing *e-h* plasma densities. In order to eliminate any role of plasma heating and carrier capture processes, the first spectra were measured at ~200–400 ps delay. Assuming the relaxation processes in QWRs are qualitatively similar to those in quantum wells (QW) then at these times following the excitation the plasma temperature will be relatively low (~50–60 K) and should vary very slowly with time; as will be shown below this assumption turns out to be fairly accurate. The clear advantages of the time resolved technique with imaging are the well defined plasma density and the low and constant lattice temperature throughout the experiment. Thus, all the spectra were taken under practically the same conditions and can be quantitatively compared.

In what follows we define the zero of the time axis as being the time after excitation at which the spectrum is characterized by a width of approximately 40 meV (at a delay of ~200 ps after excitation). In Fig. 1, we show a series of PL spectra taken at different delay times after excitation. The first three subbands can be clearly resolved at early times, which corresponds to the highest densities attained in this experiment (the rise in the PL above 1.63 eV can be attributed to the emission from the side QW that is located on both sides of the QWR [14]). A remarkable feature is that the position of the different peaks remains virtually the same with density, i.e., at different delays. In particular, there is negligible shift of the first subband peak as the second subband gradually empties. We can see a sequential emptying of the various subbands: The intensity of the PL line from the first subband remains constant while the next upper subband has not emptied substantially. This is clearly manifested in Fig. 2, which shows the constancy of the $n = 1$ transition at 1.585 eV for more than 500 ps. Also remarkable is the fact that the width and the shape of the first transition remain constant up to a density for which the intensity of the second transition begins to be sizable. It is only then that the $n = 1$ transition begins to broaden. Finally, the total width of the short time spectrum is as large as 40 meV demonstrating the very large carrier density which is excited in the wire (we estimate about $3 \times 10^6\ \mathrm{cm}^{-1}$ [23]).

It has been common practice to interpret high density spectra in quantum wires as being due to the recombination of an *e-h* plasma [12,13,15,16]. In such a framework our observations would suggest that the BGR is very small and that the recombination process in the wires preserves the *k*-selection rule up to the highest density [24] (as is the case in QWs [25]). We could also conclude, by examining the high energy tail of the spectra, that the carrier temperature is between 50 and 60 K. However, our spectra range over two orders of magnitude in density, and consequently one would expect a transition from an *e-h* plasma to a phase in which excitonic correlations start dominating. So this description is clearly inadequate, at least in the low density limit.

To address this issue we analyzed the radiative decay rate of the system. Given our experimental procedure, this rate can be directly deduced from the *time decay of the integrated luminescence,* the latter being proportional to the total density of carriers in our system. The results are plotted in Fig. 3 where it can be seen that the intensity for a QWR decays exponentially over almost two orders of magnitude, with a time constant of 560 ps. This time constant is similar to that found at much lower densities, where it corresponds to the excitonic lifetime of the wire [26]. The same behavior of a constant radiative decay rate at high and low densities is observed in various QWR samples, with different lateral subband spacing.

In order to have a more comprehensive picture we repeated the above experiments on a 100 Å multiple quantum well structure and similarly extracted the integrated

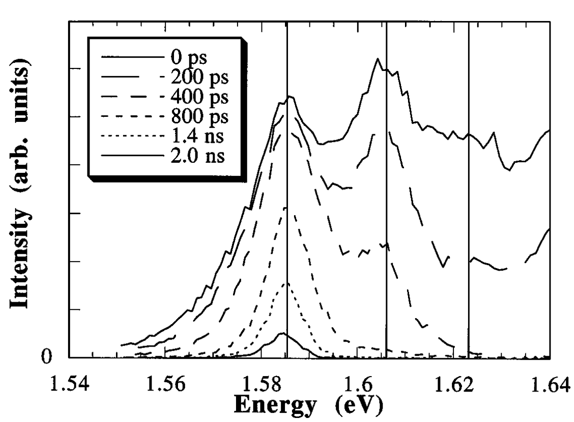

FIG. 1. High density spectra of one of the wires at successive time delays, i.e., at decreasing densities. Note the constant position of the different transitions.

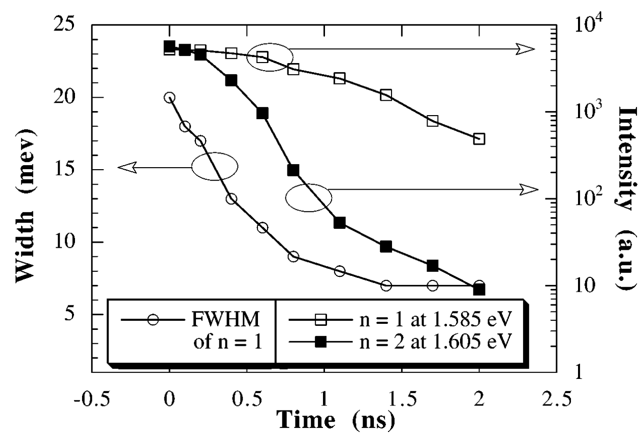

FIG. 2. Time resolved peak intensity and width of the first transition. The intensity stays absolutely constant over a very long delay indicating refilling of the $n = 1$ state which is emptied due to recombination.

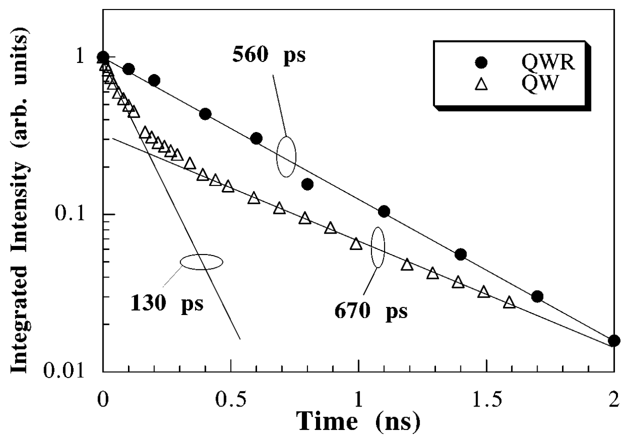

FIG. 3. Integrated luminescence intensity as a function of delay for a quantum wire and a quantum well. Note the single exponential decay over 2 orders of magnitude in the case of the wire, while for the QW there is a very distinct change of slope when the plasma becomes nondegenerate.

intensity as a function of time (see Fig. 3). The behavior of the QW is distinct from that observed in the QWR sample. Notice the clear change in the recombination rate—from a characteristic lifetime of about 130 ps at high densities to 670 ps for the excitonic luminescence—that occurs as the plasma density decreases.

To understand this fundamental difference in the behavior of QWRs and QWs, the physics of the equilibrium between excitons and a dense *e-h* plasma ($X \rightleftharpoons e + h$) needs to be introduced. In order to do this we use a well known result, the Saha equation, derived from plasma physics. Although this equation is valid only in the low density regime where Boltzmann statistics is valid, the insight it provides regarding the population of excitons on the one hand and free electrons and holes on the other, as a function of total carrier density is useful. In quantum wires the Saha equation reads

$$\frac{N_e N_h}{N_X} \approx \left(\frac{2\mu k_B T}{\pi \hbar^2}\right)^{\frac{1}{2}} \exp - \frac{E_b}{k_B T}, \tag{1}$$

where N_e, N_h, N_X are, respectively, the density of electrons, holes, and excitons, μ is the reduced mass of the electron-hole pair, T is the plasma temperature, and E_b is the exciton binding energy.

A very similar equation holds for the two dimensional case [27]. Now, one of the more paradoxical predictions of this equation is that at low carrier densities the thermodynamic equilibrium is dominated by the free electrons and holes while at higher carrier densities it is the excitons that dominate. In the case of quantum wells one may then wonder why the exact opposite behavior is observed in luminescence, i.e., excitonic recombination at low densities while at high densities the recombination spectrum is due to free electrons and holes. A detailed discussion will be given elsewhere [28] but we briefly outline the main points here. At high densities ($>10^{11}$ cm^{-2}) excitons are screened out, and it is the free carriers that govern the emission process. At low densities, on the other hand, excitons dominate the emission spectrum for although their numbers are much smaller they recombine much more efficiently than free carriers (at that density); the latter recombining via the slower bimolecular process.

For quantum wires it can be shown that the enhanced exciton binding energy shifts the equilibrium, at a given temperature, in favor of excitons. And since the importance of screening is greatly reduced in one dimension, excitonic binding and non-negligible excitonic enhancement may persist up to very high densities [3].

The next issue to consider is the relative contribution of excitons and free electrons and holes to the observed PL. It has been shown that the Sommerfeld factor in QWRs is much smaller than unity near the band edge [1–3]. This is again very different from quantum wells, where the Sommerfeld factor is larger than unity. This small value in QWRs implies that the electron-hole dipole interaction, which is proportional to the Sommerfeld factor, is reduced near the band edge. We therefore expect a substantial lowering of the recombination rate of electron-hole pairs in the continuum. This is confirmed by the absence of a series of double peaks in the PLE spectra (one due the exciton and the other to the 1D joint density of states) [20,21]. It should be noted that the above argument does not imply that only excitons exist in quantum wires at high densities. A phase space filling argument indeed limits their density to $\sim 8 \times 10^5$ cm^{-1} in each subband. However, as they occupy phase space at the bottom of the band these excitonic correlations will reduce the number of states near the band edge available to carriers in the continuum. *Both these effects thus tend to diminish the contribution of e-h pairs to the observed PL in QWRs and result in a recombination spectrum which is dominated by excitonic correlations even at the highest densities which were reached in our experiment.* Our interpretation then explains the constant lifetime of 560 ps, which cannot be understood if the plasma were composed of free electron-hole pairs. For if the luminescence in QWRs were in fact dominated by an *e-h* plasma at high densities we would also expect to see a change in the recombination time, as the density fell.

Further evidence that substantiates our hypothesis is provided by the temperature dependence of the lifetime. Repeating the measurement at higher temperature we observed an increase of the lifetime with temperature: 750 ps at 80 K and 1.2 ns at 100 K. Such a trend is consistent with excitonic luminescence and not with a degenerate plasma of electrons and holes. These experiments also tend to indicate that the carrier temperature in our sample at 8 K is indeed *below* 80 K, as otherwise there would have been no change in the radiative lifetime between the 8 and 80 K experiments.

Finally, we wish to address the issue of BGR in quantum wires. As pointed out above, if one attempts to fit the PL spectra with a free carrier model, no significant shift of the onset of the first and second subbands is obtained [29]. In fact, this is the only unambiguous result of such a fit (the fit is not unique with regards to the other parameters). Note that the constancy of the peak positions *and* the complete absence of broadening of the line for moderate densities sets very strong constraints on any fitting procedure. If, on the other hand, we consider the lowest peak as excitonic, as suggested by our data, the fact that it does not shift (or negligibly so) would mean that the BGR is exactly compensated by the reduction in exciton binding energy. This sets an upper limit of 10 to 15 meV (the exciton binding energy) for BGR at a density of about 3×10^6 cm^{-1}. Such a small value for the BGR is perfectly consistent with our claim for the existence of strong excitonic correlations up to very high densities.

In conclusion, we have studied the time resolved luminescence from highly photoexcited quantum wires. By conducting a careful experiment on high quality samples we have shown that there is no shift of the subbands energies at high densities and that the decay rate of the integrated PL intensity is constant over a large density range. Our findings suggest that the recombination spectrum is dominated by excitonic correlations even at densities that are well above the expected Mott density, and that BGR in wires is very weak even when different subbands are occupied. Our results are also of significance for the development of QWR lasers, as theoretical analysis of their properties will have to include Coulomb correlations. It has indeed been reported that lasing in wires might be due to excitons [4] and that excitonic correlations might be stable at room temperature and very large densities [3]. Our results would have to be extended to room temperature, but we may already safely state that the simple argument of the modification of the density of states cannot be used to promote 1D systems for device applications.

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