Transient carrier transfer in tunnel injection structures

V. G. Talalaev,1,2,3 J. W. Tomm1,a) N. D. Zakharov,2 P. Werner,2 U. Gösele,2 B. V. Novikov,3 A. S. Sokolov,3,4 Y. B. Samsonenko,5,6,7 V. A. Egorov,6,7 and G. E. Cirlin5,6,7

1Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, 12489 Berlin, Germany
2Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle (Saale), Germany
3Fock Institute of Physics, Saint-Petersburg State University, 198504 St. Petersburg, Russia
4Fakultät für Physik und Astronomie, Universität Heidelberg, 69120 Heidelberg, Germany
5Ioffe Physico-Technical Institute RAS, 191021 St. Petersburg, Russia
6Physics and Technology Centre for Research and Education RAS, 195220 St. Petersburg, Russia
7Institute for Analytical Instrumentation RAS, 190103 St. Petersburg, Russia

(Received 12 June 2008; accepted 3 July 2008; published online 23 July 2008)

InGaAs tunnel injection nanostructures consisting of a single quantum well as injector and a quantum dot layer as emitter are studied by time-resolved photoluminescence spectroscopy. The quantum dot photoluminescence undergoes substantial changes when proceeding from direct quantum dot excitation to quantum well excitation, which causes an indirect population of the dot ground states. This results in a lowered effective carrier temperature within the dots. Results on the carrier transfer versus barrier thickness are discussed within the Wentzel–Kramers–Brillouin approximation.

Tunnel injection structures are interesting for applications in diode lasers since they offer additional degrees of freedom for the design of the gain region. This is given by

Potential results in reduced gain suppression, increased quantum efficiency, decreased diffusion capacitance, and lower low-frequency roll-off and high-frequency chirp.1 Furthermore the expected preferential population of QDs of a particular size eventually leads to reduced inhomogeneous broadening of the gain (emission) spectrum.2 These predictions have been verified in QW- and QD-based devices that demonstrate an improved high-speed behavior; see Ref. 3 and additional references on other device implementations therein.

In this letter, we report on the analysis of the transient carrier transfer between a QW and QDs in a special type of tunnel-injection nanostructures, where QW and QDs are serving as injector and emitters, respectively. Such a structure has been suggested by Evtikhiev et al.4 First we characterize these structures and point to a unique feature of them, namely an “inverted energy band offset” within the conduction bands. Then we analyze the kinetics of the carrier transfer between the vertically stacked QW and the QDs by time-resolved photoluminescence (PL) spectroscopy by measuring a set of structures with different barrier thicknesses. Finally, we discuss the results within the framework of the Wentzel–Kramers–Brillouin (WKB) approximation and assign the observed deviations of the data from this model for barrier thicknesses <5 nm to the formation of nanobridges between QW and QDs. Evidence for the formation of such bridges is provided by transmission electron microscopy (TEM).

InGaAs tunnel injection structures are grown by solid-source molecular beam epitaxy with the following sequence of layers: InAs QDs (2 ML), GaAs spacer (thickness determined by TEM), and a single 11 nm thick In0.15Ga0.85As QW, all embedded into a GaAs matrix. This layer sequence is rather unusual and is expected to promote the formation of nanobridges. Reference samples containing either exclusively QDs or QWs are grown as well. Structural information (morphology of QD and QW, local chemical composition) was received by TEM investigation techniques (i) by diffraction contrast mode (Phillips CM20 microscope, acceleration voltage $U=200$ kV) and (ii) by image processing and analysis of high-resolution lattice structure micrographs (JEM 4010, $U=400$ kV). The In0.6Ga0.4As QDs have a diameter of 18 nm, a height of 4 nm, and an array density of $\sim5\times10^{10}$ cm$^{-2}$ (standard deviation of these parameters is $\sim10\%$). Using this information, calculations within the framework of the effective mass approximation are performed, providing a conduction band structure as follows: QWs, $E_A0=58$ meV; disk-shaped QDs, $E_A0=109$ meV and $E_A1=43$ meV. Here $E_A$ denotes the energetic distance between the quantum-confined electron state (0 or 1) and the conduction band edge of the adjacent barrier. These values have been experimentally confirmed by Arrhenius analysis of continuous wave (cw) PL intensities providing activation energies of $E_A0=55\pm3$ meV for the QW and $E_A0=105\pm7$ meV and $E_A1=35\pm2$ meV for QDs. A scheme representing these facts is depicted in Fig. 1d).

Time-resolved PL measurements are performed at 10 K with sub-100-fs excitation pulses using a mode-locked Ti:sapphire laser operating at 82 MHz. The photon energies and excitation power density are 1.33–1.60 eV and $\sim5\times10^{11}$ photons pulse$^{-1}$ cm$^{-2}$, respectively. The PL is dispersed in an imaging monochromator and detected by a synchro-scan streak camera equipped with an infrared-enhanced cathode. The time resolution of the total system is
scopic measurements are done at $T=10$ K. The spectroscopic measurements are done with a standard setup, whereas the PL excitation (PLE) experiment was done by using a KOHERAS SuperK power source.

Figure 1 provides selected results from a basic characterization of the samples. Cross-sectional dark field images of samples with barrier thickness 6.5 and 3.1 nm are shown in Figs. 1(a) and 1(b), respectively. These micrographs were taken by applying the chemically sensitive (200) reflection. Thereby In containing regions (QW and QDs) appear as darker areas. As visible in Fig. 1(a), the QW is well separated from QDs, whereas in Fig. 1(b) the QD touches the QW. This type of nanobridge is presented as an image processed high-resolution micrograph in Fig. 1(c). Inside the contour, the In content exceeds 15%, see Ref. 5. Figure 1(e) presents PL and PLE spectra from a sample with 6.5 nm barrier thickness. Tentative assignments of the optical transitions are given, with the subscripts 0 and 1 standing for ground and excited state transitions. These attributions are confirmed by both the calculations and the evolution of the PL intensities when increasing the population density by switching from cw to impulsive excitation. Figure 2 shows transient PL data. PL maps as detected by the streak camera system are shown in Figs. 2(a) and 2(b), whereas Figs. 2(c) and 2(d) show vertical cuts from maps, i.e., PL transients at particular photon energies. We compare the PL transients in the spectral channels of QW$^0$ (c) and QD$^0$ (d) ground state recombination, respectively, for two different samples with barrier widths of 3.1 nm (black full circles) and 6.5 nm (red open circles) barrier thickness. Figure 3 summarizes the tunneling
times as derived from such transient PL data measured from the whole sample set.

It is worthwhile to address the implications of our results. First, we point to the band structure. According to Fig. 1(e) the spectral positions of the QW ground state and the QD excited and ground state transitions are at about 1.36, 1.27, and 1.2 eV, respectively. According to calculations and Arrhenius analysis, the electron states (in the conduction band) do not follow this order (on the absolute energy scale); see Fig. 1(d). Figures 2(a) and 2(b) show the result that proves this tentative assignment. The only difference between both measurements is an increase of the excitation photon energy by 90 meV from 1.33 to 1.42 eV. For an excitation energy below the QW absorption edge, we observe a substantial increase of the QD \( \sigma \) luminescence (decay time \( \sim 750 \) ps); see Fig. 2(b). The increased QD \( \sigma \) PL is likely to be caused by the efficient population of the electronic QD ground states by tunnel injection from the QWs. Thus we show that in this particular structure pumping at increased photon energies results in “colder” luminescence. The term colder points here to the fact that the ratio of the populations of ground and excited states is altered in favor of the ground state population. Basically, the result shown in Figs. 2(a) and 2(b) is a clear demonstration of the functionality of a tunnel injection structure. This is also a necessary condition for the following analysis of the PL transients.

Tunneling times \( \tau \) are obtained by fitting transient PL curves such as shown in Figs. 2(c) and 2(d). Both the QW PL decays and the QDs PL rises are impacted by \( \tau \), since they involve the depopulation of the injector and the population of the emitters, respectively. The actual transients, however, are formed additionally by the recombination lifetime \( \tau_{QW} \), and \( \tau_{QD} \), which are determined separately at the reference samples to 420 and 750 ps, respectively. Thus the transient depleting of the QW ground state is described by two competing processes and produces a QW PL decay curve \( I_{QW} \) that is shaped according to

\[
I_{QW}(t) = I_{max} \exp\left(-\frac{t}{\tau_{QW}}\right) \exp\left(-\frac{t}{\tau}\right).
\]

(1)

Here \( I_{max} \) corresponds to the maximum of the profile.

The QD PL rise is well-described by two noncompeting processes. Taking into account the instantaneous population, the rise is described by

\[
I_{QD}(t) = I_0 \exp\left(-\frac{t}{\tau_{QD}}\right) + \frac{I_{max} \times \tau_{QD}}{\tau_{QD} - \tau} \left[\exp\left(-\frac{t}{\tau_{QD}}\right) - \frac{t}{\tau}\right],
\]

(2)

where \( I_0 \) corresponds to the intensity where the excitation pulse peaks.

Thus there are two independent PL data sets, namely QW PL decay and QD PL rise, which provide information on \( \tau \). Therefore the excellent agreement between the two obtained \( \tau \)-data sets (see circles and squares in Fig. 3) provides additional evidence for the supposed carrier transfer scenario. On the other hand the actual values of \( \tau_{QW} \), as well as the rise time of the QD PL set limits for the applicability of the two approaches; see open circles in Fig. 3. All these results are summarized in Fig. 3 together with \( \tau \) values that have been obtained for the carrier transfer between differently sized QDs.\(^*\)\(^*\) As long as the barrier thickness exceeds about 6 nm, remarkable agreement with the data from Ref. 6 and the WKB approximation is visible. For this thickness range, we also do not find any systematic dependence of QD-transition energies on the barrier thickness. For lower barrier thicknesses, however, our actual \( \tau \)-values fall substantially shorter and approach the time resolution limits of our setup. We assume that the significant reduction of the transfer time is explained by the formation of point-contact-like channels (nanobridges) between the apexes of some QDs and the QW. The high resolved TEM image of a structure with a barrier width of 3.1 nm, see Fig. 1(e), shows such an contact nanobridge with a width of about 1 nm. These nanobridges are expected to form channels that allow for ultrafast carrier exchange between QDs and the QW. A very similar type of such contacts was previously observed for closely stacked InAs QD layers.\(^*\)

A complementary analysis of the tunnel-injection mechanism as well as the determination of reliable \( \tau \)-values for the barrier thickness range below 5 nm, in particular in presence of nanobridges, represent tasks for ongoing investigations.

Summarizing, we report on the transient carrier transfer in tunnel-injection nanostructures containing a QW and a QD layer, which are coupled through a barrier. A set of structures with barrier thickness in the 2–9.5 nm range is analyzed. For QW excitation substantial cooling of the carriers confined within the QDs is demonstrated, compared to the situation when the QDs are populated exclusively by their own absorption. The carrier transfer from the QW into QDs is measured and the results are discussed within the framework of the WKB approximation for tunneling. Observed deviations from this model for barrier thicknesses <5 nm are assigned to the formation of nano-bridges across the tunneling barriers. Evidence for the existence of such bridges is provided by TEM analysis.

Helpful discussion with M. Ziegler and expert technical assistance by R. Hoffmann is gratefully acknowledged. This work has been supported by the Russian Foundation for Basic Research (08-02-00954) and Program RAS “Quantum Nanostructures.”


