Picosecond Spin-Preserving Carrier Capture in InGaAs/GaAs Quantum Dots

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Summary. Carrier capture into self-organized InGaAs/GaAs quantum dots is studied in a femtosecond transmission experiment. Resonantly generating carriers in the wetting layer, we analyze the population of both the band edge of the wetting layer and the quantum dot excited states. We find a carrier capture time of 3 ps that is independent of the carrier density providing that it remains small compared to the number of available electronic states. Moreover, we find that the capture process is predominantly spin-preserving in nature. These results suggest that phonon mediated scattering governs the quantum dot filling.

Semiconductor quantum dots (QDs) are attractive both as model systems to study the physics of zero-dimensional structures and for their device applications. In particular, the discrete energy level structure of QDs offers significant advantages for modern semiconductor laser technology. In QD based devices, nonequilibrium carrier dynamics plays a central role in determining the performance limitations. As a result, especially intra-QD relaxation processes have been subject to a considerable research effort [1-7]. However, the important initial step of the population transfer from the two dimensional wetting layer (WL) into the zero-dimensional structures has not been isolated experimentally and remains subject of lively discussions [8-10].

In this contribution, we directly analyze the transfer of photoinjected carriers from the WL to fully quantized QD levels. The experiment relies on the transient bleaching of the WL band edge and the QD interband transitions in a femtosecond transmission experiment. For low excitation densities, we find a capture time of 3 ps indicating a very efficient QD filling. Most interestingly, the capture time does not

significantly depend on the density of carriers photoinjected into the WL. Moreover, exploiting the selection rules for circularly polarized light, the carrier capture of electrons is shown to be spin-preserving.

The nanostructures investigated in this study are self-organized InGaAs QDs grown on a GaAs. The ground state transitions of the inhomogeneously broadened ensemble are centered at 1.25 eV and the interband energetic spacing of the transitions from different QD shells is approximately 40 meV, i.e. close to the optical phonon energies of GaAs The scheme of our experimental approach is displayed in the inset of Fig. 1(a). A pump pulse with a duration of 100 fs is tuned to the absorption continuum of the WL, thus resonantly creating electron-hole pairs. After relaxation towards the band edge, the carriers are transfered into fully quantized QD levels. Correspondingly, a transient optical bleaching signal is established reflecting the occupation of the electronic states. These transmission changes of the QD monolayer are detected with a 20 fs broadband probe pulse, that is derived from the second branch of a two color-Ti:sapphire laser, is spectrally dispersed after transmission through the specimen and detected with a photodiode.

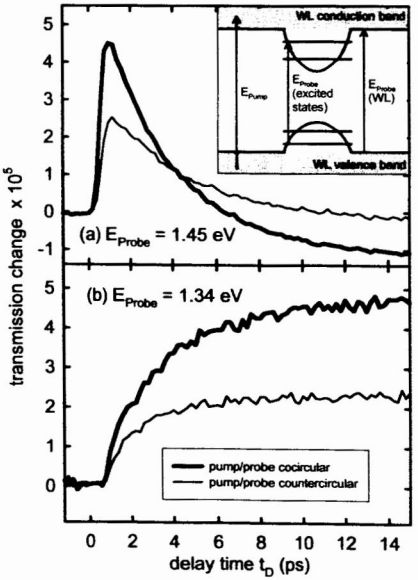

Fig. 1 (a) Transmission changes of the QD monolayer detected at the band edge of the WL after resonant carrier injection at $t_D = 0$ for co- and countercircular polarization of the excitation and probe pulses and a temperature of T = 4 K. The inset sketches the bandstructure with the vertical arrows indicating the interband transitions of the study. (b) Corresponding transmission changes probing the excited states of the QDs.

Results for a moderate excitation density and a probe photon energy of 1.45 eV, near the band edge of the WL, are shown in Fig. 1(a). The bleaching signal decays nicely exponential with a time constant of 3 ps. These dynamics are accompanied by the buildup of a transmission change

probing the QD excited levels (see Fig. 1(b)). Thus, the present measurement directly traces the population transfer from the two dimensional WL into the QDs.

More detailed insight into the carrier capture process is gained exploiting the spin selectivity of the interband transitions. Circularly polarized excitation yields a preferential spin orientation of the photogenerated carriers. While the spin orientation of holes is rapidly destroyed, the electron spin in the two-dimensional WL is expected to be preserved over the timescale of the present study. As a consequence, detecting the transmission changes of the WL with a counter-circularly polarized probe pulse yields an overall reduction of the transmission change by a factor of two (see thin line in Fig. 1(a)). More surprisingly, the nonlinear optical response of the QD interband transitions depends on the polarization configuration in the same manner as observed for the WL (see Fig. 1(b)). These observations demonstrate that the carrier capture process is a predominantly spin-preserving process.

In order to identify the nature of the capture process, the QD filling is studied for various excitation densities. To this end, we analyze the transient transmission changes of the WL band edge at a probe photon energy of 1.45 eV as a function of the excitation power. For weak excitation, we find that an exponential decay with a time constant of 3 ps provides a good fit to the experimental data. Most interestingly, the decay times extracted from exponential fits (see filled circles in Fig. 2) do not significantly depend on the excitation density indicating that carrier-carrier scattering has only a minor influence on the capture dynamics.

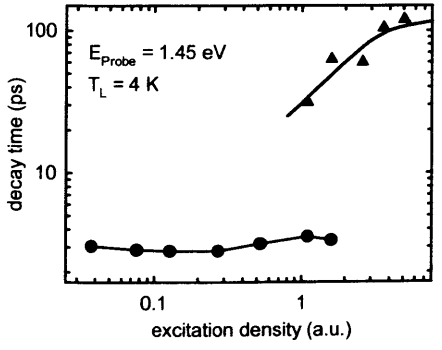

Fig.2 Decay times of the WL population for various excitation densities. The time constants are extracted from the transmission changes of the WL band edge at 1.45~eV by single and double exponential fits as discussed in the text. The abscissa may be interpreted as an estimate for the number of photogenerated electron-hole pairs per QD.

Increasing the excitation density to values comparable or larger than the QD density on the sample, the decay of the bleaching signal is effectively slowed down due to the emergence of a slower signal component. This

finding is readily understood from the onset of macroscopic state filling effects in the QDs. Modelling these dynamics with a bi-exponential fit, we identify a fast capture process with a time constant of again 3 ps and a relatively long-lived component (see the filled triangles in Fig. 2). For the highest excitation densities of the study, the signal is dominated by massive QD filling and decays only on a timescale approaching typical radiative multiexciton lifetimes in QDs.

The independence of the capture time on the excitation density clearly points towards a phonon mediated process. It is interesting to relate this finding to previous experimental results claiming the importance of carrier-carrier scattering for the relaxation dynamics [1,3,4]. These time-resolved luminescence experiments are intrinsically sensitive to a combination of carrier capture and relaxation towards the QD ground state. In contrast, we study the capture into electronic states near the onset of the WL continuum. Our results are supported by an ultrafast luminescence study on large InGaAs quantum dots with a comparable level spacing [5].

In conclusion, we have analyzed the ultrafast, spin-preserving population transfer from a two dimensional WL to fully quantized QD levels in a femtosecond transmission experiment. The carrier capture time in our QDs with a level spacing comparable or smaller as the optical phonon energies amounts to 3 ps and is found to be independent of the excitation density. Taken together, these observations strongly indicate a phonon mediated process. As a result, we see no significant phonon bottleneck for the filling of the QD states. This finding may be an important ingredient for the optimization of modern QD based laser devices.

References

1. B. Ohnesorge, et al., Phys. Rev. B **54**, 11532 (1996)
2. R. Heitz, et al., Phys. Rev. B **56**, 10435 (1997)
3. M. De Giorgi, et al., Appl. Phys. Lett. **79**, 3968 (2001)
4. D. Morris, et al., Appl. Phys. Lett. **75**, 3593 (1999)
5. T. F. Boggess, et al., Appl. Phys. Lett. **78**, 276 (2001)
6. T. S. Sosnowski, et al., Phys. Rev. B **57**, R9423 (1998)
7. Müller, et al., Appl. Phys. Lett. **83**, 3572 (2003)
8. U. Bockelmann, and G. Bastard, Phys. Rev. B **42**, 8947 (1990)
9. H. Benisty, et al., Phys. Rev. B **44**, 10945 (1991)
10. T. R. Nielsen, et al., Phys. Rev. B **69**, 235314