10th Int. Symp. "Nanostructures: Physics and Technology" QWR/QD.06
St Petersburg, Russia, June 17-21, 2002

Optical memory concepts with self-organized quantum dots — material systems and energy-selective charging

C. Kapteyn†, J. Ehehalt†, R. Heitz†, D. Bimberg†, G. E. Cirlin‡, V. M. Ustinov‡ and N. N. Ledentsov†‡

† Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstra\sse 36, 10623 Berlin, Germany
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. For memory structures based on optically induced charge in self-organized quantum dots, the concept of wavelength-domain multiplexing in the quantum dot ensemble is an essential prerequisite. The electric properties of quantum dots in various material systems as studied by time-resolved capacitance spectroscopy are summarized, and candidates suitable for future memory applications are discussed. By combining optical excitation and capacitance spectroscopy, direct evidence is obtained for energy-selective hole charge generation and storage in InAs/GaAs quantum dots. A clear dependence of the activation energy of the emitted holes on the energy of the excitation is observed.

Introduction

The ultimate reduction of device size and charge in semiconductor memory devices naturally leads to the quantum regime. An ideal prototype to study few- and single-electron effects are semiconductor quantum dot heterostructures [1]. Early it was realized that self-organized quantum dots can be fabricated in a variety of material systems with a resulting broad range of electric and optical properties. As direct electrical addressing of huge ensembles of such quantum dots would involve enormous technological efforts, it was proposed to employ optical addressing to circumvent these difficulties [2,3]. Optical addressing is feasible only provided wavelength-domain multiplexing can be realized. Self-organized quantum dots exhibit a natural spectral distribution in their energy levels due to ensemble fluctuations in size and shape. They can also be fabricated with very high optical quality and very low defect density. Such quantum dot heterostructures are therefore a very promising system for studying wavelength-domain multiplexing techniques. Optical charging in suitable sample structures with quantum dots embedded has recently been reported [3–8]. A demonstration, however, that the generated charge retains the initial spectral information of the excitation process is, although crucial, still lacking.

Here, we discuss the prerequisites for memory applications based on self-organized quantum dots and study the optical charge generation process by combining optical charging with conventional capacitance spectroscopy. Our experimental results demonstrate the feasibility of energy-selective optical charging of sub-ensembles of quantum dots.

Material systems

Central issues for memory concepts are the energy level structure, the carrier retention time, and the escape mechanisms. A simple diode structure allows to directly detect the amount of charge in an embedded quantum dot layer, which is situated at a suitable distance from a Schottky- or pn-interface. Such a structure is an ideal device to study charging and escape processes in quantum dots with help of time-resolved capacitance spectroscopy. Based on the data evaluation methods of deep level transient spectroscopy (DLTS) [9], capacitance transients are recorded, emission time constants determined, escape mechanisms identified, and activation energies derived.

A variety of material combinations exhibiting quantum dot formation have so far been investigated [10-17]. The obtained activation energies are summarized in Table 1. For memory applications a high activation energy is desirable, since thermal escape of carriers is usually the limiting process for the charge storage time.

Table Activation energies obtained from time-resolved capacitance measurements for various quantum dot material systems.

Material system	Carrier type	Activation energy	Reference
InAs/GaAs	electron	80–90 meV (*)	[11,12]
InAs/GaAs	hole	160–250 meV	[12,17]
GaSb/GaAs	hole	350–400 meV	[14,16]
InAs/InP	hole	440 meV	[15]
InP/GaInP	electron	220 meV	[10]
Ge/Si	hole	350 meV	[13]

From the experimental results one can estimate room temperature carrier retention times by extrapolation (e.g. a few ps for holes in InAs/GaAs quantum dots [12] and about one μs for holes in Ge/Si). These retention times are significantly shorter than retention times, which can be attained in suitable memory devices, where carrier storage of up to several seconds was observed [3–7]. The reduced retention times in time-resolved capacitance measurements are due to the presence of an electric field, which increases the effective emission rate. Typical memory structures make use of an optimized band structure and additional barriers in order to prevent unintentional carrier loss. Table 1 nevertheless clearly indicates in which material systems long intrinsic storage times may be expected.

From this point of view, hole storage in Ge/Si, InAs/InP, or GaSb/GaAs quantum dots appears promising.

For the employment of wavelength-domain multiplexing techniques the optical properties of the material system are of great importance, too. As GaSb/GaAs has a type-II band line-up, only the hole states are fully quantized in all three dimensions and a true energy-selectivity might hence not be given for the absorption process. For a general test of the feasibility of the concept of optical multiplexing, InAs/GaAs quantum dots hence appear to be a very promising system. In previous experiments it was furthermore observed, that electron and hole escape processes, and hence the according emission rates in the InAs/GaAs material system, are significantly different [12]. This is reflected by the smaller thermal activation energy for electrons. A net hole charge can thus elegantly be created, simply by optical excitation of electron-hole pairs and fast release of electrons. Due to these reasons, we decided to focus on InAs/GaAs quantum dots for optical charging experiments.

Optical multiplexing

For optical addressing of sub-ensembles of self-organized quantum dots one can employ their natural fluctuations in the optical transition energies. Due to the quantum-size effect, larger quantum dots exhibit a smaller transition energy, whereas smaller quantum dots show a larger transition energy. Conclusively one can expect to charge large quantum dots by illumination at a lower energy than small quantum dots, which require a larger excitation energy. In order to avoid additional absorption in impurities or defects, and carrier loss due to charge-transfer into defects close to the quantum dots, a high material quality is required.

Experimental details

Based on time-resolved capacitance measurements, we have performed experiments with optical excitation. The sample structures are GaAs p^+n-diodes similar to devices previously used for purely electrical capacitance spectroscopy of quantum dots, see e.g. [11]. Details of the sample structure and the optical charging process will be presented elsewhere [17].

In our experiments, we record capacitance transients due to carrier emission from the quantum dots after illumination of the sample with monochromatic light with a resolution of about 6 meV for 10 ms. A conventional lamp with a monochromator was employed for these measurements. The temperature shift of the absorption was accounted for by adjusting the actual excitation wavelength according to the temperature. Here and in the following, only the respective excitation energies for 300 K are mentioned.

Results

Recording capacitance transients after illumination, a minority carrier DLTS signal is clearly observed (not shown). From the amplitude a hole charge of about 0.2 per quantum

dot is deduced. The activation energy of the thermally activated emission process exhibits a pronounced dependence on the energy of the exciting light. The reference sample did not exhibit any optically induced DLTS signal. Bias dependent DLTS measurements of the quantum dot sample (not shown) prove that the quantum dot layer is the source of the observed carrier emission.

By increasing the excitation energy in the range between 1.02 eV and 1.09 eV, a shifting DLTS signal with an accordingly decreasing activation energy down to about 180 meV is observed, see Fig. 1. For further increasing excitation energy, the activation energy quickly raises again to about 230 meV at around 1.11 eV, until it decreases again down to about 170 meV for the highest excitation energies (up to 1.19 eV).

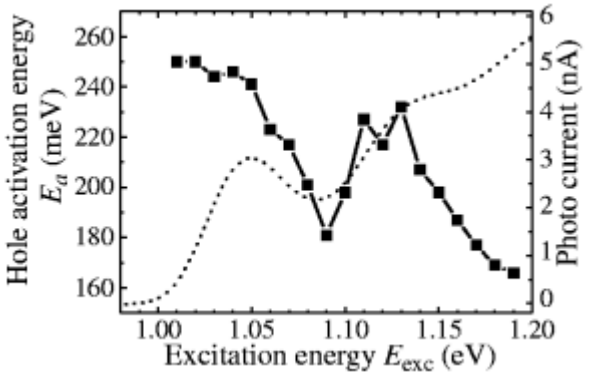

Fig 1. The dependence of the hole activation energy as function of excitation energy (for 300 K) is shown as squares. The reverse bias was set to 8.3 V and optical pulses of 10 ms were applied. For comparison, the photo-absorption current at 300 K for a reverse bias of 8.3 V is displayed as dotted line.

These experimental findings are interpreted in terms of energy-selective charging of sub-ensembles of quantum dots by resonant absorption of light. In large quantum dots with a low ground state transition energy a net hole charge is generated, which exhibits a large activation energy. By increasing the excitation energy, smaller and smaller quantum dots are resonantly charged with holes, which are energetically closer to the GaAs valence band edge and hence lead to smaller activation energies. This process is even repeated for absorption in the first excited state, as the observed activation energies for excitation above around 1.11 eV reflect. These experimental results demonstrate, to our knowledge, the first direct proof for energy-selective charging of quantum dot sub-ensembles.

Concluding remarks

In order to charge quantum dots and hence "write" data in future memory structures, optical addressing is a very promising approach. Various quantum dot material systems have previously been studied by time-resolved capacitance spectroscopy. By comparing these results and also taking optical properties into account InAs/GaAs was identified as a promising candidate for optical charging experiments. By combining resonant optical excitation and capacitance spectroscopy, it is demonstrated that optical multiplexing in quantum dot ensembles is feasible. Hole ground state activation energies in the range of 250 meV to 180 meV were observed for excitation energies between 1.02 eV and 1.09 eV, giving evidence for energy-selective charging. Similar energy-selective charging could alse be conducted for optical charging of excited quantum dot states.

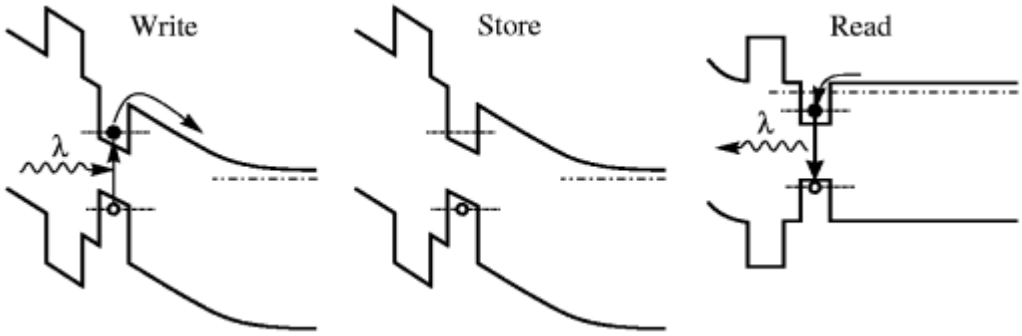

Fig 2. Schematic band structure and basic operation principle of a memory structure based on energy-selective optical charging, similar to the sample investigated here. Note the additional barriers for increased hole storage times.

The development of memory structures based on optical multiplexing in self-organized quantum dot ensembles is still at a rather explorative stage. Recent results from the investigation of the electrical properties and carrier emission processes in quantum dots in various material systems, however, allow to conduct more dedicated research towards key functional ingredients of future devices, especially optical addressing.

In the future it is intended to increase the spectral resolution, to extend the investigations to other promising material systems (e.g. GaSb/GaAs), and to develop and study optimized sample structures for memory applications, which allow for increased carrier retention times and hence more realistic device operation.

Acknowledgements

This work was funded by the NANOMAT project of the European Commission's Growth Programme, contract number G5RD-CT-2001-00545, and SFB 296 of DFG.

References

1. D. Bimberg, M. Grundmann and N. N. Ledentsov, *Quantum Dot Heterostructures*, John Wiley & Sons, Chichester (1998)
2. S. Muto, *Jpn. J. Appl. Phys.* **34**, L210 (1995).
3. K. Imamura, Y. Sugiyama, Y. Nakata et al, *Jpn. J. Appl. Phys.* **34**, L1445 (1995).
4. G. Yusa and H. Sakaki, *Appl. Phys. Lett.* **70**, 345 (1997).
5. J. J. Finley, M. Skalitz, M. Arzberger et al, *Appl. Phys. Lett.* **73**, 2618 (1998).
6. Y. Sugiyama, Y. Nakata, S. Muto et al, *IEEE J. Sel. Topics in Quantum Electron.* **4**, 880 (1998).
7. T. Lundstrom, W. Schoenfeld, H. Lee et al, *Science* **286**, 2312 (1999).
8. H. Pettersson, L. Baath, N. Carlson et al, *Appl. Phys. Lett.* **79**, 78 (2001).
9. D. V. Lang, *J. Appl. Phys.* **45**, 3023 (1974).
10. S. Anand, N. Carlsson, M.-E. Pistol et al, *J. Appl. Phys.* **84**, 3747 (1998).
11. C. M. A. Kapteyn, F. Heinrichsdorff, O. Stier et al, *Phys. Rev. B* **60**, 14265 (1999).

12. C. M. A. Kapteyn, M. Lion, R. Heitz et al, *Appl. Phys. Lett.* **76**, 1573 (2000).
13. C. M. A. Kapteyn, M. Lion, R. Heitz et al, *Appl. Phys. Lett.* **77**, 4169 (2000).
14. R. Magno, B. R. Bennett and E. R. Glaser, *J. Appl. Phys.* **88**, 5843 (2000).
15. H. Pettersson, C. Pryor, L. Landin et al, *Phys. Rev. B* **61**, 4795 (2000).
16. M. Geller et al, *to be published*
17. C. Kapteyn et al, *to be published*
