Optical spectroscopy of carrier dynamics in semiconductor nanostructures

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Summary

In the thesis, entitled “Optical spectroscopy of carrier dynamics in semiconductor nanostructures”, the PhD research on the light-matter interactions in several low-dimensional semiconductor structures is presented. It comprises an elaborate study of the spectral and temporal characteristics of recombination and relaxation processes of excited carriers in nanostructures of Si, GaN and perovskites to gain insights on the properties of these materials with the aim to develop more efficient optoelectronic devices.

The first Chapter introduces some important concepts for the rest of the thesis. Fundamental optical properties of bulk semiconductors and possible ways to overcome their limitations for applications in optoelectronics are discussed. Two potential ways for improvement are elaborated upon: down-scaling the material dimensions toward the material-specific exciton Bohr radius (for most materials between $\sim$1$-$50nm), allowing quantum confinement effects to modify the properties, and doping the material with rare earth ions to which energy transfer can take place. Throughout the whole thesis both possibilities are extensively discussed. Specific carrier dynamics processes, which are thoroughly investigated in this research, are described and the Chapter ends with a short description of all the Chapters.

In Chapter 2, nanocrystals of Si, one of, or possibly even the most important semiconductor material in nowadays society, are discussed as a possible solution to overcome the fundamental efficiency limit for photovoltaic conversion (the Shockley-Queisser limit). Large losses appear on both sides of the solar spectrum through lack of absorption of low-energy photons and only partial use of high-energy photons due to fast thermalisation. Several approaches to achieve efficient spectral conversion and their feasibilities are presented.

A power-dependent study of photoluminescence properties of Si nanocrystals embedded into an SiO_2 matrix is presented in Chapter 3. In contrast to pulsed excitation, the photoluminescence under continuous wave excitation does not saturate, as expected due to efficient non-radiative Auger recombination under high excitation power. The experiments demonstrate that through laser-induced heating, which is especially effective under continuous wave excitation, the radiative (photon) emission rate can be enhanced. This finding provides a possible avenue to enhance the optical faculty of Si and could also be relevant for the use of Si nanocrystals in future photovoltaic applications.

In Chapter 4, the ultrafast carrier dynamics of the system of solid-state dispersions of Si nanocrystals in an SiO_2 matrix is investigated with transient induced absorption spectroscopy. The spectral dependence of the free-carrier dynamics of the Si nanocrystals in an oxygen-rich environment is explained with the formation of the self-trapped exciton state on the surface of the nanocrystal. Supported with theoretical modeling, the results provide new insights into the self trapping of free excitons on the surface-related states, which is found to be dependent on the nanocrystal size, and could be of importance to enhance the optical performance of this material.

The optical properties of slightly larger freestanding faceted Si crystalline nanoparticles, too large for quantum confinement effects to play a role, are described in Chapter 5. These nanoparticles feature a superlinear flux dependence of the photoluminescence intensity following a power-law with exponents up to $\sim$10. Nanoparticles of different sizes and shapes are investigated and the effect of phosphorous doping is also considered, indicating that the porosity of the layer is linked to the photoluminescence intensity. Through Raman spectroscopy it is revealed that, as for the Si nanocrystals described in Chapter 3, the local temperature can increase significantly under continuous wave laser illumination. It is postulated that due to the reduced heat conductivity, the Si grains can be laser-heated resulting in the strong emission.

The next Chapter is devoted to optical doping of an SiO_2 matrix with Si nanocrystals and Er (Chapter 6). The effect of Er doping on the free-carrier dynamics is studied by transient induced spectroscopy

and the results are discussed together with the photoluminescence dynamics, to investigate the energy transfer mechanisms between the Si nanocrystals and the Er^{3+} ions. Specific emphasis is on the fast energy transfer from the Si nanocrystals to the lowest excited state of the Er^{3+} ion.

Chapter 7 also focuses on optical doping and energy transfer between a semiconductor and rare earth ions. A different system, namely the wide-bandgap semiconductor GaN doped with Eu, which is seen as a promising alternative material for light-emitting devices, is the topic of research in this Chapter. Photoluminescence and transient induced absorption measurements are performed on pure and Eu-doped GaN layers. Determination of the ultrafast carrier dynamics is complicated, and some issues and solutions are presented.

The last Chapter deals with all-inorganic cesium lead halide perovskite nanocrystals, which are recently synthesized and now widely investigated. This material shows very promising properties for a wide range of applications. In the first part of the Chapter, the multiexciton lifetime is determined through fluence-dependent transient induced absorption spectroscopy. This lifetime is of crucial importance for the photoluminescence properties under high excitation power, which are investigated and cross-correlated with induced absorption measurements in the second part of the Chapter, but also for the investigations of the photoluminescence quantum yield and carrier multiplication. Through this multiplication process multiple carriers can be generated upon absorption of a single (high-energy) photon. This carrier multiplication process and a specific form, called space-separated quantum cutting, are discussed in the last part of the Chapter.

Samenvatting

In het proefschrift, getiteld "Optical spectroscopy of carrier dynamics in semiconductor nanostructures", wordt het promotieonderzoek over de interactie tussen licht en materie van verschillende laagdimensionale halfgeleiderstructuren gepresenteerd. Het omvat een uitgebreide studie van de spectrale en tijdsgerelateerde karakteristieken van recombinatie- en relaxatieprocessen van aangeslagen ladingsdragers in nanostructuren van Si, GaN en perovskieten om inzicht te krijgen in de eigenschappen van deze materialen, waarbij het ontwikkelen van efficiëntere optoelektronische apparaten een belangrijk doel is.

Het eerste Hoofdstuk introduceert een aantal belangrijke concepten voor de rest van het proefschrift. Fundamentele optische eigenschappen van bulk halfgeleiders en mogelijke manieren om hun beperkingen voor toepassingen in de optoelektronica te overwinnen worden besproken. Twee potentiële manieren voor verbetering worden uitgewerkt: het verkleinen van de materiaaldimensies richting de materiaalspecifieke exciton Bohr straal (voor de meeste materialen $\sim$1$-$50nm), waardoor kwantumbegrenzingseffecten ("quantum confinement effects") de eigenschappen kunnen veranderen, en het doteren van het materiaal met zeldzame aardmetalen waarnaar energie overgedragen kan worden. Beide mogelijkheden komen in het gehele proefschrift uitgebreid aan de orde. Specifieke tijdsafhankelijke processen van de ladingsdragers, die in dit onderzoek grondig worden onderzocht, worden beschreven en het Hoofdstuk eindigt met een korte beschrijving van alle Hoofdstukken.

In Hoofdstuk 2 worden nanokristallen van Si, een van de, of mogelijk zelfs dè belangrijkste halfgeleider in de hedendaagse maatschappij, besproken als een mogelijke oplossing om de fundamentele fotovoltaïsche omzettingsefficiëntielimiet te overwinnen (de Shockley-Queisser limiet). Grote verliezen vinden plaats aan beide kanten van het zonnespectrum door gebrek aan absorptie van laagenergetische fotonen en alleen gedeeltelijke gebruik van de hoogenergetische fotonen als gevolg van snelle thermalisatie. Verschillende benaderingen voor een efficiënte spectrale omzetting en hun haalbaarheid worden gepresenteerd.

De afhankelijkheid van de excitatie intensiteit op de fotoluminescentie eigenschappen van Si nanokristallen ingebed in een SiO_2 matrix worden gepresenteerd in Hoofdstuk 3. In tegenstelling tot gepulste excitatie, verzadigt de fotoluminescentie niet onder continue excitatie, zoals verwacht op grond van efficiënte niet-stralende Auger recombinatie bij een hoge excitatie intensiteit. De experimenten laten zien dat door lasergeïnduceerde verwarming, die vooral effectief is onder continue excitatie, de stralende fotonemissiesnelheid kan worden verhoogd. Deze bevinding biedt een mogelijke route om het optische vermogen van Si te verbeteren en kan ook relevant zijn voor het gebruik van Si nanokristallen in toekomstige fotovoltaïsche toepassingen.

In Hoofdstuk 4 worden ultrasnelle tijdsafhankelijke processen van de ladingdragers van een systeem bestaande uit dispersies van Si nanokristallen in een SiO_2 matrix onderzocht met tijdsgerelateerde geïnduceerde absorptiespectroscopie. De spectrale afhankelijkheid van de tijdsgerelateerde processen van de vrije ladingsdragers in de Si nanokristallen in een zuurstofrijke omgeving worden verklaard met de formatie van een "self-trapped exciton" toestand op de oppervlakte van het nanokristal. De resultaten worden ondersteund met theoretische modellen en verstrekken nieuwe inzichten in het "self-trapping" proces van vrije excitonen. Dit proces is afhankelijk van de grootte van het nanokristal. Deze bevindingen kunnen van belang zijn voor de verbetering van de optische prestatie van dit materiaal.

Optische eigenschappen van iets grotere vrijstaande gefacetteerde Si kristallijne nanodeeltjes, die te groot zijn om door kwantumbegrenzingseffecten te worden beïnvloed, worden beschreven in Hoofdstuk 5. Deze nanodeeltjes worden gekenmerkt door een superlineaire afhankelijkheid van invallende fotonflux op de fotoluminescentie intensiteit: een machtsfunctie met machten tot $\sim$10. Nanodeeltjes van een aantal groottes en vormen zijn onderzocht, alsmede het effect van fosfor dotering is bestudeerd. De resultaten geven aan dat de poreusheid van de laag nanodeeltjes is gekoppeld aan de fotoluminescentie intensiteit.

Door middel van Raman spectroscopie is duidelijk geworden dat, net zoals voor de Si nanokristallen die in Hoofdstuk 3 worden beschreven, de lokale temperatuur significant stijgt onder continue laserilluminatie. Verondersteld wordt dat de Si nanodeeltjes als gevolg van de verminderde warmtegeleiding kunnen worden opgewarmd door een laser, hetgeen resulteert in sterke emissie.

Het volgende Hoofdstuk is gewijd aan de dotering van een SiO_2 matrix met Si nanokristallen en Er (Hoofdstuk 6). Het effect van dotering met Er op de tijdsafhankelijke processen van vrije ladingdragers is bestudeerd met tijdsgerelateerde geïnduceerde absorptie en tijdsgerelateerde fotoluminescentie. Ze worden samen besproken om de overdracht van energie tussen de Si nanokristallen en de Er^{3+} ionen te onderzoeken. De nadruk ligt vooral op de snelle energieoverdracht van de Si nanokristallen naar de laagst aangeslagen energietoestand van het Er^{3+} ion.

Hoofdstuk 7 focust ook op de dotering en energieoverdracht tussen een halfgeleider en een zeldzaam aardmetaalion. Een ander systeem, namelijk de halfgeleider GaN gedoteerd met Eu, dat wordt gezien als een veelbelovend alternatief materiaal voor de lichtemitterende diode (LED), is het onderzoeksonderwerp van dit Hoofdstuk. Fotoluminescentie en tijdsgerelateerde geïnduceerde absorptiemetingen zijn verricht op zuivere en met Eu gedoteerde GaN lagen. Het bepalen van de ultrasnelle tijdsafhankelijke processen van de ladingsdragers is gecompliceerd, en enkele problemen en oplossingen worden gepresenteerd.

Het laatste Hoofdstuk behandelt anorganische nanokristallen van cesium lood halide perovskieten, welke sinds kort gesynthetiseerd kunnen worden en nu op grote schaal worden onderzocht. Dit materiaal laat veelbelovende eigenschappen zien voor een breed scala aan toepassingen. In het eerste deel van het Hoofdstuk, wordt de levensduur van het multiexciton bepaald met lichtintensiteitafhankelijke transiënt geïnduceerde absorptiespectroscopie. Deze levensduur is van cruciaal belang voor de fotoluminescentie eigenschappen bij hoge excitatievermogens, die worden onderzocht en kruisgecorreleerd met geïnduceerde absorptiemetingen in het tweede deel van het Hoofdstuk, maar ook voor onderzoek naar de fotoluminescentie kwantumopbrengst ("quantum yield") en het vermenigvuldigen van ladingsdragers ("carrier multiplication"). Tijdens dit laatstgenoemde vermenigvuldigingsproces kunnen meerdere ladingsdragers worden gegenereerd na absorptie van één (hoogenergetisch) foton. Dit ladingdragersvermenigvuldigingsproces en een specifieke vorm daarvan, genaamd "ruimtelijk gescheiden kwantum knippen" ("space-separated quantum cutting"), worden besproken in het laatste deel van het Hoofdstuk.

List of publications

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1. W.D.A.M. de Boer, E.M.L.D. de Jong, D. Timmerman, T. Gregorkiewicz, H. Zhang, W.J. Buma, A.N. Poddubny, A.A. Prokofiev and I.N. Yassievich. *Carrier dynamics in Si nanocrystals in an SiO_2 matrix investigated by transient light absorption*, Physical Review B 88(15):155304, 2013.

2. E.M.L.D. de Jong, S. Saeed and T. Gregorkiewicz. *Photon management using Si nanocrystals and Er^{3+} ions: Generation of hot carriers upon absorption of low-energy photons*, ECS Transactions 61(5):127, 2014 - Chapter 2.

3. S. Saeed, E.M.L.D. de Jong, K. Dohnalová and T. Gregorkiewicz. *Efficient optical extraction of hot-carrier energy*, Nature Communications 5:4665, 2014.

4. E.M.L.D. de Jong, S. Saeed and T. Gregorkiewicz. *Hoe “hete” elektronen de zonnecelefficiëntie kunnen verhogen*, Nederlands Tijdschrift voor de Natuurkunde 81(1):12, 2015.

5. S. Saeed, E.M.L.D. de Jong and T. Gregorkiewicz. *Step-like increase of quantum yield of 1.5 μm Er-related emission in SiO_2 doped with Si nanocrystals*, Journal of Applied Physics 117(6):064303, 2015.

6. Y. Ding, F. Wu, Y. Zhang, X. Liu, E.M.L.D. de Jong, T. Gregorkiewicz, X. Hong, Y. Liu, M.C.G. Aalders, W.J. Buma and H. Zhang. *Interplay between static and dynamic energy transfer in biofunctional upconversion nanoplatforms*, The Journal of Physical Chemistry Letters 6(13):2518, 2015.

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8. E.M.L.D. de Jong, G. Mannino, A. Alberti, R. Ruggeri, M. Italia, F. Zontone, Y. Chushkin, A.R. Pennisi, T. Gregorkiewicz and G. Faraci. *Strong infrared photoluminescence in highly porous layers of large faceted Si crystalline nanoparticles*, Scientific Reports 6:25644, 2016 - Chapter 5.

9. E.M.L.D. de Jong, G. Yamashita, L. Gomez, M. Ashida, Y. Fujiwara and T. Gregorkiewicz. *Multiexciton lifetime in all-inorganic $CsPbBr_3$ perovskite nanocrystals*, The Journal of Physical Chemistry C 121(3):1941, 2016 - Chapter 8.

10. E.M.L.D. de Jong, W.D.A.M. de Boer, I.N. Yassievich and T. Gregorkiewicz. *Trapping time of excitons in Si nanocrystals embedded in a SiO_2 matrix*, Physical Review B 95(19):195312, 2017 - Chapter 4.

11. E.M.L.D. de Jong, H. Rutjes, J. Valenta, M.T. Trinh, A.N. Poddubny, I.N. Yassievich, A. Capretti and T. Gregorkiewicz. *Thermally stimulated exciton emission in Si nanocrystals*, under review - Chapter 3.

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13. E.M.L.D. de Jong et al. *Energy transfer dynamics in SiO_2:Er sensitized with Si nanocrystals (working title)*, in preparation - Chapter 6.

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