



UvA-DARE (Digital Academic Repository)

Optical spectroscopy of carrier dynamics in semiconductor nanostructures

de Jong, E.M.L.D.

[Link to publication](#)

Citation for published version (APA):

de Jong, E. M-L. D. (2017). Optical spectroscopy of carrier dynamics in semiconductor nanostructures

General rights

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <http://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

Bibliography

- [1] W. Shockley and H.J. Queisser. Detailed balance limit of efficiency of p-n junction solar cells. *Journal of Applied Physics*, 32(3):510–519, 1961.
- [2] S. Richard, F. Aniel, and G. Fishman. Energy-band structure of Ge, Si, and GaAs: A thirty-band $\mathbf{k}\cdot\mathbf{p}$ method. *Physical Review B*, 70(23):235204, 2004.
- [3] A.G. Cullis, L.T. Canham, and P.D.J. Calcott. The structural and luminescence properties of porous silicon. *Journal of Applied Physics*, 82(3):909–965, 1997.
- [4] W.D.A.M. de Boer, D. Timmerman, K. Dohnalová, I.N. Yassievich, H. Zhang, W.J. Buma, and T. Gregorkiewicz. Red spectral shift and enhanced quantum efficiency in phonon-free photoluminescence from silicon nanocrystals. *Nature Nanotechnology*, 5(12):878–884, 2010.
- [5] A.J. Nozik. Spectroscopy and hot electron relaxation dynamics in semiconductor quantum wells and quantum dots. *Annual Review of Physical Chemistry*, 52(1):193–231, 2001.
- [6] A. Pandey and P. Guyot-Sionnest. Slow electron cooling in colloidal quantum dots. *Science*, 322(5903):929–932, 2008.
- [7] U. Bockelmann and G. Bastard. Phonon scattering and energy relaxation in two-, one-, and zero-dimensional electron gases. *Physical Review B*, 42(14):8947, 1990.
- [8] T. Inoshita and H. Sakaki. Electron-phonon interaction and the so-called phonon bottleneck effect in semiconductor quantum dots. *Physica B: Condensed Matter*, 227(1):373–377, 1996.
- [9] A.J. Nozik. Multiple exciton generation in semiconductor quantum dots. *Chemical Physics Letters*, 457(1):3–11, 2008.
- [10] K. Dohnalová, A.N. Poddubny, A.A. Prokofiev, W.D.A.M. de Boer, C.P. Umesh, J.M.J. Paulusse, H. Zuilhof, and T. Gregorkiewicz. Surface brightens up Si quantum dots: direct bandgap-like size-tunable emission. *Light: Science & Applications*, 2(1):e47, 2013.
- [11] L. Protesescu, S. Yakunin, M.I. Bodnarchuk, F. Krieg, R. Caputo, C.H. Hendon, R.X. Yang, A. Walsh, and M.V. Kovalenko. Nanocrystals of cesium lead halide perovskites (CsPbX_3 , $\text{X}=\text{Cl}$, Br , and I): novel optoelectronic materials showing bright emission with wide color gamut. *Nano Letters*, 15(6):3692–3696, 2015.
- [12] V.I. Klimov, A.A. Mikhailovsky, D.W. McBranch, C.A. Leatherdale, and M.G. Bawendi. Quantization of multiparticle Auger rates in semiconductor quantum dots. *Science*, 287(5455):1011–1013, 2000.
- [13] V.A. Kharchenko and M. Rosen. Auger relaxation processes in semiconductor nanocrystals and quantum wells. *Journal of Luminescence*, 70(1-6):158–169, 1996.
- [14] V.I. Klimov. Spectral and dynamical properties of multiexcitons in semiconductor nanocrystals. *Annual Review of Physical Chemistry*, 58:635–673, 2007.
- [15] D. Timmerman, I. Izeddin, and T. Gregorkiewicz. Saturation of luminescence from Si nanocrystals embedded in SiO_2 . *Physica Status Solidi A*, 207(1):183–187, 2010.
- [16] M. Wolf, R. Brendel, J.H. Werner, and H.J. Queisser. Solar cell efficiency and carrier multiplication in $\text{Si}_{1-x}\text{Ge}_x$ alloys. *Journal of Applied Physics*, 83(8):4213–4221, 1998.
- [17] O. Christensen. Quantum efficiency of the internal photoelectric effect in silicon and germanium. *Journal of Applied Physics*, 47(2):689–695, 1976.
- [18] A.J. Nozik. Nanoscience and nanostructures for photovoltaics and solar fuels. *Nano Letters*, 10(8):2735–2741, 2010.
- [19] M.C. Beard, A.G. Midgett, M.C. Hanna, J.M. Luther, B.K. Hughes, and A.J. Nozik. Comparing multiple exciton generation in quantum dots to impact ionization in bulk semiconductors: implications for enhancement of solar energy conversion. *Nano Letters*, 10(8):3019–3027, 2010.
- [20] M. Govoni, I. Marri, and S. Ossicini. Carrier multiplication between interacting nanocrystals for fostering silicon-based photovoltaics. *Nature Photonics*, 6(10):672–679, 2012.
- [21] D. Timmerman, I. Izeddin, P. Stallinga, I.N. Yassievich, and T. Gregorkiewicz. Space-separated quantum cutting with silicon nanocrystals for photovoltaic applications. *Nature Photonics*, 2(2):105–109, 2008.

- [22] M.T. Trinh, R. Limpens, W.D.A.M. de Boer, J.M. Schins, L.D.A. Siebbeles, and T. Gregorkiewicz. Direct generation of multiple excitons in adjacent silicon nanocrystals revealed by induced absorption. *Nature Photonics*, 6(5):316–321, 2012.
- [23] G.B. Hazel, J.B. Hedrick, G.J. Orris, P.H. Stauffer, and J.W. Hendley II. Rare earth elements: critical resources for high technology. *USGS Fact Sheet*, 087-02, 2002.
- [24] O. Semonin, J.M. Luther, and M.C. Beard. Multiple exciton generation in a quantum dot solar cell. *SPIE (International Society for Optics and Photonics) Newsroom*, 2012.
- [25] S. Rühle. Tabulated values of the Shockley–Queisser limit for single junction solar cells. *Solar Energy*, 130:139–147, 2016.
- [26] R.R. King, D. Bhusari, D. Larrabee, X.Q. Liu, E. Rehder, K. Edmondson, H. Cotal, R.K. Jones, J.H. Ermer, C.M. Fetzer, D.C. Law, and N.H. Karam. Solar cell generations over 40% efficiency. *Progress in Photovoltaics: Research and Applications*, 20(6):801–815, 2012.
- [27] M.A. Green. *Third generation photovoltaics*. Springer, 2006.
- [28] F. Priolo, T. Gregorkiewicz, M. Galli, and T.F. Krauss. Silicon nanostructures for photonics and photovoltaics. *Nature Nanotechnology*, 9(1):19–32, 2014.
- [29] R.T. Wegh, H. Donker, K.D. Oskam, and A. Meijerink. Visible quantum cutting in $\text{LiGdF}_4:\text{Eu}^{3+}$ through downconversion. *Science*, 283(5402):663–666, 1999.
- [30] A.J. Nozik. Quantum dot solar cells. *Physica E: Low-dimensional Systems and Nanostructures*, 14(1):115–120, 2002.
- [31] R.D. Schaller, M. Sykora, J.M. Pietryga, and V.I. Klimov. Seven excitons at a cost of one: redefining the limits for conversion efficiency of photons into charge carriers. *Nano Letters*, 6(3):424–429, 2006.
- [32] M.C. Beard, K.P. Knutsen, P. Yu, J.M. Luther, Q. Song, W.K. Metzger, R.J. Ellingson, and A.J. Nozik. Multiple exciton generation in colloidal silicon nanocrystals. *Nano Letters*, 7(8):2506–2512, 2007.
- [33] C. Smith and D. Binks. Multiple exciton generation in colloidal nanocrystals. *Nanomaterials*, 4(1):19–45, 2013.
- [34] D. Timmerman, J. Valenta, K. Dohnalová, W.D.A.M. de Boer, and T. Gregorkiewicz. Step-like enhancement of luminescence quantum yield of silicon nanocrystals. *Nature Nanotechnology*, 6(11):710–713, 2011.
- [35] M.T. Trinh, R. Limpens, and T. Gregorkiewicz. Experimental investigations and modeling of Auger recombination in silicon nanocrystals. *The Journal of Physical Chemistry C*, 117(11):5963–5968, 2013.
- [36] W.D.A.M. de Boer, D. Timmerman, T. Gregorkiewicz, H. Zhang, W.J. Buma, A.N. Poddubny, A.A. Prokofiev, and I.N. Yassievich. Self-trapped exciton state in Si nanocrystals revealed by induced absorption. *Physical Review B*, 85(16):161409(R), 2012.
- [37] A. Brewer and K. von Haefen. In situ passivation and blue luminescence of silicon clusters using a cluster beam/ H_2O codeposition production method. *Applied Physics Letters*, 94(26):261102, 2009.
- [38] L. Tsybeskov, J.V. Vandyshev, and P.M. Fauchet. Blue emission in porous silicon: Oxygen-related photoluminescence. *Physical Review B*, 49(11):7821, 1994.
- [39] 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.
- [40] G.S. He, Q. Zheng, K.T. Yong, F. Erogbogbo, M.T. Swihart, and P.N. Prasad. Two- and three-photon absorption and frequency upconverted emission of silicon quantum dots. *Nano Letters*, 8(9):2688–2692, 2008.
- [41] R.T. Ross and A.J. Nozik. Efficiency of hot-carrier solar energy converters. *Journal of Applied Physics*, 53(5):3813–3818, 1982.
- [42] D. König, K. Casalenuovo, Y. Takeda, G. Conibeer, J.F. Guillemoles, R. Patterson, L.M. Huang, and M.A. Green. Hot carrier solar cells: Principles, materials and design. *Physica E: Low-dimensional Systems and Nanostructures*, 42(10):2862–2866, 2010.
- [43] A.A. Prokofiev, A.N. Poddubny, and I.N. Yassievich. Phonon decay in silicon nanocrystals: Fast phonon recycling. *Physical Review B*, 89(12):125409, 2014.

- [44] I. Izeddin, A.S. Moskalenko, I.N. Yassievich, M. Fujii, and T. Gregorkiewicz. Nanosecond dynamics of the near-infrared photoluminescence of Er-doped SiO₂ sensitized with Si nanocrystals. *Physical Review Letters*, 97(20):207401, 2006.
- [45] I. Izeddin, D. Timmerman, T. Gregorkiewicz, A.S. Moskalenko, A.A. Prokofiev, I.N. Yassievich, and M. Fujii. Energy transfer in Er-doped SiO₂ sensitized with Si nanocrystals. *Physical Review B*, 78(3):035327, 2008.
- [46] S. Saeed, D. Timmerman, and T. Gregorkiewicz. Dynamics and microscopic origin of fast 1.5 μm emission in Er-doped SiO₂ sensitized with Si nanocrystals. *Physical Review B*, 83(15):155323, 2011.
- [47] N.N. Ha, S. Cueff, K. Dohnalová, M.T. Trinh, C. Labbé, R. Rizk, I.N. Yassievich, and T. Gregorkiewicz. Photon cutting for excitation of Er³⁺ ions in SiO₂ sensitized by Si quantum dots. *Physical Review B*, 84(24):241308, 2011.
- [48] 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 SiO₂ doped with Si nanocrystals. *Journal of Applied Physics*, 117(6):064303, 2015.
- [49] 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.
- [50] R. Limpens. *Carrier dynamics in coupled silicon nanocrystal systems*. PhD thesis, University of Amsterdam, 2016.
- [51] W.L. Wilson, P.F. Szajowski, and L.E. Brus. Quantum confinement in size-selected, surface-oxidized silicon nanocrystals. *Science*, 262:1242–1242, 1993.
- [52] D. Kovalev, H. Heckler, G. Polisski, and F. Koch. Optical properties of Si nanocrystals. *Physica Status Solidi B*, 215(2):871–932, 1999.
- [53] M. Sykora, L. Mangolini, R.D. Schaller, U. Kortshagen, D. Jurbergs, and V.I. Klimov. Size-dependent intrinsic radiative decay rates of silicon nanocrystals at large confinement energies. *Physical Review Letters*, 100(6):067401, 2008.
- [54] A.J. Minnich, M.S. Dresselhaus, Z.F. Ren, and G. Chen. Bulk nanostructured thermoelectric materials: current research and future prospects. *Energy & Environmental Science*, 2(5):466–479, 2009.
- [55] M. Achermann, A.P. Bartko, J.A. Hollingsworth, and V.I. Klimov. The effect of Auger heating on intraband carrier relaxation in semiconductor quantum rods. *Nature Physics*, 2(8):557–561, 2006.
- [56] R. Limpens, A. Lesage, M. Fujii, and T. Gregorkiewicz. Size confinement of Si nanocrystals in multi-nanolayer structures. *Scientific Reports*, 5:17289, 2015.
- [57] J. Valenta, M. Greben, S. Gutsch, D. Hiller, and M. Zacharias. Effects of inter-nanocrystal distance on luminescence quantum yield in ensembles of Si nanocrystals. *Applied Physics Letters*, 105(24):243107, 2014.
- [58] A.M. Hartel, D. Hiller, S. Gutsch, P. Löper, S. Estradé, F. Peiró, B. Garrido, and M. Zacharias. Formation of size-controlled silicon nanocrystals in plasma enhanced chemical vapor deposition grown SiO_xN_y/SiO₂ superlattices. *Thin Solid Films*, 520(1):121–125, 2011.
- [59] M. Lax. Temperature rise induced by a laser beam. *Journal of Applied Physics*, 48(9):3919–3924, 1977.
- [60] C. Delerue, M. Lannoo, G. Allan, E. Martin, I. Mihalcescu, J.C. Vial, R. Romestain, F. Muller, and A. Bsiesy. Auger and Coulomb charging effects in semiconductor nanocrystallites. *Physical Review Letters*, 75(11):2228, 1995.
- [61] D. Timmerman and T. Gregorkiewicz. Power-dependent spectral shift of photoluminescence from ensembles of silicon nanocrystals. *Nanoscale Research Letters*, 7(1):389, 2012.
- [62] D. Kovalev, J. Diener, H. Heckler, G. Polisski, N. Künzner, and F. Koch. Optical absorption cross sections of Si nanocrystals. *Physical Review B*, 61(7):4485, 2000.
- [63] 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*, 6:25664, 2016.
- [64] G. Faraci, A.R. Pennisi, A. Alberti, R. Ruggeri, and G. Mannino. Giant photoluminescence emission in crystalline faceted Si grains. *Scientific Reports*, 3:2674, 2013.
- [65] V. Poborchii, T. Tada, and T. Kanayama. Giant heating of Si nanoparticles by weak laser light: Optical microscopic study and application to particle modification. *Journal of Applied Physics*, 97(10):104323, 2005.

- [66] L. Han, M. Zeman, and A.H.M. Smets. Raman study of laser-induced heating effects in free-standing silicon nanocrystals. *Nanoscale*, 7(18):8389–8397, 2015.
- [67] H. Koyama and P.M. Fauchet. Laser-induced thermal effects on the optical properties of free-standing porous silicon films. *Journal of Applied Physics*, 87(4):1788–1794, 2000.
- [68] H. Koyama and P.M. Fauchet. Very large continuous-wave-laser-induced optical absorption in porous silicon films: Evidence for thermal effects. *Applied Physics Letters*, 73(22):3259–3261, 1998.
- [69] M.J. Konstantinović, S. Bersier, X. Wang, M. Hayne, P. Lievens, R.E. Silverans, and V.V. Moshchalkov. Raman scattering in cluster-deposited nanogranular silicon films. *Physical Review B*, 66(16):161311, 2002.
- [70] S.K. Estreicher, T.M. Gibbons, B. Kang, and M.B. Bebek. Phonons and defects in semiconductors and nanostructures: Phonon trapping, phonon scattering, and heat flow at heterojunctions. *Journal of Applied Physics*, 115(1):012012, 2014.
- [71] G. Davies. The optical properties of luminescence centres in silicon. *Physics Reports*, 176(3-4):83–188, 1989.
- [72] T. Trupke, M.A. Green, P. Würfel, P.P. Altermatt, A. Wang, J. Zhao, and R. Corkish. Temperature dependence of the radiative recombination coefficient of intrinsic crystalline silicon. *Journal of Applied Physics*, 94(8):4930–4937, 2003.
- [73] W. Gerlach, H. Schlangenotto, and H. Maeder. On the radiative recombination rate in silicon. *Physica Status Solidi A*, 13(1):277–283, 1972.
- [74] A.S. Moskalenko, J. Berakdar, A.A. Prokofiev, and I.N. Yassievich. Single-particle states in spherical Si/SiO₂ quantum dots. *Physical Review B*, 76(8):085427, 2007.
- [75] A.D. Yoffe. Low-dimensional systems: quantum size effects and electronic properties of semiconductor microcrystallites (zero-dimensional systems) and some quasi-two-dimensional systems. *Advances in Physics*, 42(2):173–262, 1993.
- [76] J.H. Lienhard IV and V. Lienhard. *A heat transfer textbook*. Phlogiston Press, 2008.
- [77] C.H. Henager and W.T. Pawlewicz. Thermal conductivities of thin, sputtered optical films. *Applied Optics*, 32(1):91–101, 1993.
- [78] F.R. Brotzen, P.J. Loos, and D.P. Brady. Thermal conductivity of thin SiO₂ films. *Thin Solid Films*, 207(1-2):197–201, 1992.
- [79] F. Pevere, I. Sychugov, F. Sangghaleh, A. Fucikova, and J. Linnros. Biexciton emission as a probe of Auger recombination in individual silicon nanocrystals. *The Journal of Physical Chemistry C*, 119(13):7499–7505, 2015.
- [80] I. Mihalcescu, J.C. Vial, A. Bsiesy, F. Muller, R. Romestain, E. Martin, C. Delerue, M. Lannoo, and G. Allan. Saturation and voltage quenching of porous-silicon luminescence and the importance of the Auger effect. *Physical Review B*, 51(24):17605, 1995.
- [81] N.V. Kurova and V.A. Burdov. Resonance structure of the rate of Auger recombination in silicon nanocrystals. *Semiconductors*, 44(11):1414–1417, 2010.
- [82] F. Trojánek, K. Neudert, M. Bittner, and P. Malý. Picosecond photoluminescence and transient absorption in silicon nanocrystals. *Physical Review B*, 72(7):075365, 2005.
- [83] K. Ueda, T. Tayagaki, M. Fukuda, M. Fujii, and Y. Kanemitsu. Breakdown of the k-conservation rule in quantized Auger recombination in Si_{1-x}Ge_x nanocrystals. *Physical Review B*, 86(15):155316, 2012.
- [84] M.R. Bergren, P.K.B. Palomaki, N.R. Neale, T.E. Furtak, and M.C. Beard. Size-dependent exciton formation dynamics in colloidal silicon quantum dots. *ACS Nano*, 10(2):2316–2323, 2016.
- [85] J.A. McGuire, J. Joo, J.M. Pietryga, R.D. Schaller, and V.I. Klimov. New aspects of carrier multiplication in semiconductor nanocrystals. *Accounts of Chemical Research*, 41(12):1810–1819, 2008.
- [86] Y.S. Park, A.V. Malko, J. Vela, Y. Chen, Y. Ghosh, F. García-Santamaría, J.A. Hollingsworth, V.I. Klimov, and H. Htoon. Near-unity quantum yields of biexciton emission from CdSe/CdS nanocrystals measured using single-particle spectroscopy. *Physical Review Letters*, 106(18):187401, 2011.
- [87] L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franzó, and F. Priolo. Optical gain in silicon nanocrystals. *Nature*, 408(6811):440–444, 2000.
- [88] O. Boyraz and B. Jalali. Demonstration of a silicon Raman laser. *Optics Express*, 12(21):5269–5273, 2004.

- [89] E.M.L.D. de Jong, S. Saeed, W.C. Sinke, and T. Gregorkiewicz. Generation of hot carriers for photon management in future photovoltaics. *Solar Energy Materials and Solar Cells*, 135:67–71, 2015.
- [90] J.H. Park, L. Gu, G. von Maltzahn, E. Ruoslahti, S.N. Bhatia, and M.J. Sailor. Biodegradable luminescent porous silicon nanoparticles for in vivo applications. *Nature Materials*, 8(4):331–336, 2009.
- [91] M.V. Wolkin, J. Jorne, P.M. Fauchet, G. Allan, and C. Delerue. Electronic states and luminescence in porous silicon quantum dots: the role of oxygen. *Physical Review Letters*, 82(1):197, 1999.
- [92] G. Allan, C. Delerue, and M. Lannoo. Nature of luminescent surface states of semiconductor nanocrystallites. *Physical Review Letters*, 76(16):2961, 1996.
- [93] A.V. Gert and I.N. Yassievich. Role of surface self-trapped excitons in the energy relaxation of photoexcited silicon nanocrystals. *Semiconductors*, 49(4):492–497, 2015.
- [94] S. Takeoka, M. Fujii, and S. Hayashi. Size-dependent photoluminescence from surface-oxidized Si nanocrystals in a weak confinement regime. *Physical Review B*, 62(24):16820, 2000.
- [95] F. Trojánek, K. Neudert, P. Malý, K. Dohnalová, and I. Pelant. Ultrafast photoluminescence in silicon nanocrystals studied by femtosecond up-conversion technique. *Journal of Applied Physics*, 99:116108, 2006.
- [96] A.N. Poddubny, A.A. Prokofiev, and I.N. Yassievich. Optical transitions and energy relaxation of hot carriers in Si nanocrystals. *Applied Physics Letters*, 97(23):231116, 2010.
- [97] H. Bethe and R. Peierls. Quantum theory of the dipton. *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, 148(863):146–156, 1935.
- [98] D. Babić, R. Tsu, and R.F. Greene. Ground-state energies of one- and two-electron silicon dots in an amorphous silicon dioxide matrix. *Physical Review B*, 45(24):14150, 1992.
- [99] R. Claps, D. Dimitropoulos, V. Raghunathan, Y. Han, and B. Jalali. Observation of stimulated Raman amplification in silicon waveguides. *Optics Express*, 11(15):1731–1739, 2003.
- [100] G. Faraci, S. Gibilisco, A.R. Pennisi, G. Franzó, S. La Rosa, and L. Lozzi. Catalytic role of adsorbates in the photoluminescence emission of Si nanocrystals. *Physical Review B*, 78(24):245425, 2008.
- [101] G. Mannino, A. Alberti, R. Ruggeri, S. Libertino, A.R. Pennisi, and G. Faraci. Octahedral faceted Si nanoparticles as optical traps with enormous yield amplification. *Scientific Reports*, 5:8354, 2015.
- [102] M. Zebarjadi, K. Esfarjani, M.S. Dresselhaus, Z.F. Ren, and G. Chen. Perspectives on thermoelectrics: from fundamentals to device applications. *Energy & Environmental Science*, 5(1):5147–5162, 2012.
- [103] G. Faraci, S. Gibilisco, and A.R. Pennisi. Superheating of silicon nanocrystals observed by Raman spectroscopy. *Physics Letters A*, 373(41):3779–3782, 2009.
- [104] Y. Kanemitsu. Efficient light emission from crystalline and amorphous silicon nanostructures. *Journal of Luminescence*, 100(1):209–217, 2002.
- [105] G. Faraci, G. Mannino, A.R. Pennisi, R. Ruggeri, P. Sberna, and V. Privitera. Raman and photoluminescence spectroscopy of Si nanocrystals: Evidence of a form factor. *Journal of Applied Physics*, 113(6):063518, 2013.
- [106] G. Faraci, S. Gibilisco, and A.R. Pennisi. Quantum confinement and thermal effects on the Raman spectra of Si nanocrystals. *Physical Review B*, 80(19):193410, 2009.
- [107] V. Alex, S. Finkbeiner, and J. Weber. Temperature dependence of the indirect energy gap in crystalline silicon. *Journal of Applied Physics*, 79(9):6943–6946, 1996.
- [108] P. Lautenschlager, P.B. Allen, and M. Cardona. Temperature dependence of band gaps in Si and Ge. *Physical Review B*, 31(4):2163, 1985.
- [109] J. Miao, P. Charalambous, J. Kirz, and D. Sayre. Extending the methodology of X-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens. *Nature*, 400(6742):342–344, 1999.
- [110] A.S. Barnard and P. Zapol. A model for the phase stability of arbitrary nanoparticles as a function of size and shape. *The Journal of Chemical Physics*, 121(9):4276–4283, 2004.
- [111] U. Kortshagen. Nonthermal plasma synthesis of semiconductor nanocrystals. *Journal of Physics D: Applied Physics*, 42(11):113001, 2009.
- [112] Y. Chushkin, F. Zontone, E. Lima, L. De Caro, P. Guardia, L. Manna, and C. Giannini. Three-dimensional coherent diffractive imaging on non-periodic specimens at the ESRF beamline ID10. *Journal of Synchrotron Radiation*, 21(3):594–599, 2014.

- [113] K.W. Böer. *Survey of Semiconductor Physics: Volume II Barriers, Junctions, Surfaces, and Devices*. Springer Science & Business Media, 2012.
- [114] D. König, S. Gutsch, H. Gnaser, M. Wahl, M. Kopnarski, J. Göttlicher, R. Steininger, M. Zacharias, and D. Hiller. Location and electronic nature of phosphorus in the Si nanocrystal- SiO₂ system. *Scientific Reports*, 5:9702, 2015.
- [115] S.K. Gupta and P.K. Jha. Modified phonon confinement model for size dependent Raman shift and linewidth of silicon nanocrystals. *Solid State Communications*, 149(45):1989–1992, 2009.
- [116] K. Wang, R.W. Martin, K.P. O'Donnell, V. Katchkanov, E. Nogales, K. Lorenz, E. Alves, S. Ruffenach, and O. Briot. Selectively excited photoluminescence from Eu-implanted GaN. *Applied Physics Letters*, 87(11):112107, 2005.
- [117] 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 nanoplateforms. *The Journal of Physical Chemistry Letters*, 6(13):2518–2523, 2015.
- [118] M. Miritello, R.L. Savio, P. Cardile, and F. Priolo. Enhanced down conversion of photons emitted by photoexcited Er_xY_{2-x}Si₂O₇ films grown on silicon. *Physical Review B*, 81(4):041411, 2010.
- [119] W.J. Miniscalco. Erbium-doped glasses for fiber amplifiers at 1500 nm. *Journal of Lightwave Technology*, 9(2):234–250, 1991.
- [120] D. Pacifici, G. Franzò, F. Priolo, F. Iacona, and L. Dal Negro. Modeling and perspectives of the Si nanocrystals–Er interaction for optical amplification. *Physical Review B*, 67(24):245301, 2003.
- [121] H. Ennen, J. Schneider, G. Pomrenke, and A. Axmann. 1.54- μ m luminescence of erbium-implanted III-V semiconductors and silicon. *Applied Physics Letters*, 43(10):943–945, 1983.
- [122] A.J. Kenyon, C.E. Chryssou, C.W. Pitt, T. Shimizu-Iwayama, D.E. Hole, N. Sharma, and C.J. Humphreys. Luminescence from erbium-doped silicon nanocrystals in silica: Excitation mechanisms. *Journal of Applied Physics*, 91(1):367–374, 2002.
- [123] P.G. Kik, M.J.A. de Dood, K. Kikoin, and A. Polman. Excitation and deexcitation of Er³⁺ in crystalline silicon. *Applied Physics Letters*, 70(13):1721–1723, 1997.
- [124] F. Priolo, G. Franzò, S. Coffa, and A. Carnera. Excitation and nonradiative deexcitation processes of Er³⁺ in crystalline Si. *Physical Review B*, 57(8):4443, 1998.
- [125] D. Timmerman. *Optical spectroscopy of carrier multiplication by silicon nanocrystals*. PhD thesis, University of Amsterdam, 2012.
- [126] X.L. Wu, Y.F. Mei, G.G. Siu, K.L. Wong, K. Moulding, M.J. Stokes, C.L. Fu, and X.M. Bao. Spherical growth and surface-quasifree vibrations of Si nanocrystallites in Er-doped Si nanostructures. *Physical Review Letters*, 86(14):3000, 2001.
- [127] F. Priolo, G. Franzò, F. Iacona, D. Pacifici, and V. Vinciguerra. Excitation and non-radiative de-excitation processes in Er-doped Si nanocrystals. *Materials Science and Engineering: B*, 81(1):9–15, 2001.
- [128] G. Franzó, V. Vinciguerra, and F. Priolo. The excitation mechanism of rare-earth ions in silicon nanocrystals. *Applied Physics A*, 69(1):3–12, 1999.
- [129] P.G. Kik and A. Polman. Exciton–erbium interactions in Si nanocrystal-doped SiO₂. *Journal of Applied Physics*, 88(4):1992–1998, 2000.
- [130] M. Fujii, M. Yoshida, Y. Kanzawa, S. Hayashi, and K. Yamamoto. 1.54 μ m photoluminescence of Er³⁺ doped into SiO₂ films containing Si nanocrystals: evidence for energy transfer from Si nanocrystals to Er³⁺. *Applied Physics Letters*, 71(9):1198–1200, 1997.
- [131] R. Berera, R. van Grondelle, and J.T.M. Kennis. Ultrafast transient absorption spectroscopy: principles and application to photosynthetic systems. *Photosynthesis Research*, 101(2-3):105–118, 2009.
- [132] W.D.A.M. de Boer, M.T. Trinh, D. Timmerman, J.M. Schins, L.D.A. Siebbeles, and T. Gregorkiewicz. Increased carrier generation rate in Si nanocrystals in SiO₂ investigated by induced absorption. *Applied Physics Letters*, 99(5):053126, 2011.
- [133] I. Robel, R. Gresback, U. Kortshagen, R.D. Schaller, and V.I. Klimov. Universal size-dependent trend in Auger recombination in direct-gap and indirect-gap semiconductor nanocrystals. *Physical Review Letters*, 102(17):177404, 2009.

- [134] P.N. Favennec, H. L'haridon, M. Salvi, D. Moutonnet, and Y. Le Guillou. Luminescence of erbium implanted in various semiconductors: IV, III-V and II-VI materials. *Electronics Letters*, 25(11):718–719, 1989.
- [135] Y. Jiang, Y. Li, Y. Li, Z. Deng, T. Lu, Z. Ma, P. Zuo, L. Dai, L. Wang, H. Jia, W. Wang, J. Zhou, W. Liu, and H. Chen. Realization of high-luminous-efficiency InGaN light-emitting diodes in the “green gap” range. *Scientific Reports*, 5:10883, 2015.
- [136] A. Kaneta, M. Funato, and Y. Kawakami. Nanoscopic recombination processes in InGaN/GaN quantum wells emitting violet, blue, and green spectra. *Physical Review B*, 78(12):125317, 2008.
- [137] M.K. Horton, S. Rhode, S.L. Sahonta, M.J. Kappers, S.J. Haigh, T.J. Pennycook, C.J. Humphreys, R.O. Dusane, and M.A. Moram. Segregation of In to dislocations in InGaN. *Nano Letters*, 15(2):923–930, 2015.
- [138] L.T. Romano, M.D. McCluskey, C.G. Van de Walle, J.E. Northrup, D.P. Bour, M. Kneissl, T. Suski, and J. Jun. Phase separation in InGaN multiple quantum wells annealed at high nitrogen pressures. *Applied Physics Letters*, 75(25):3950–3952, 1999.
- [139] A. Nishikawa, N. Furukawa, T. Kawasaki, Y. Terai, and Y. Fujiwara. Improved luminescence properties of Eu-doped GaN light-emitting diodes grown by atmospheric-pressure organometallic vapor phase epitaxy. *Applied Physics Letters*, 97(5):051113, 2010.
- [140] Y.Q. Wang and A.J. Steckl. Three-color integration on rare-earth-doped GaN electroluminescent thin films. *Applied Physics Letters*, 82(4):502–504, 2003.
- [141] W.D.A.M. de Boer, C. McGonigle, T. Gregorkiewicz, Y. Fujiwara, S. Tanabe, and P. Stallinga. Optical excitation and external photoluminescence quantum efficiency of Eu^{3+} in GaN. *Scientific Reports*, 4:5235, 2014.
- [142] N. Furukawa, A. Nishikawa, T. Kawasaki, Y. Terai, and Y. Fujiwara. Atmospheric pressure growth of Eu-doped GaN by organometallic vapor phase epitaxy. *Physica Status Solidi A*, 208(2):445–448, 2011.
- [143] J.F. Muth, J.H. Lee, I.K. Shmagin, R.M. Kolbas, H.C. Casey Jr, B.P. Keller, U.K. Mishra, and S.P. DenBaars. Absorption coefficient, energy gap, exciton binding energy, and recombination lifetime of GaN obtained from transmission measurements. *Applied Physics Letters*, 71(18):2572–2574, 1997.
- [144] R. Wakamatsu, D.G. Lee, A. Koizumi, V. Dierolf, Y. Terai, and Y. Fujiwara. Luminescence properties of Eu-doped GaN grown on GaN substrate. *Japanese Journal of Applied Physics*, 52(8S):08JM03, 2013.
- [145] S. Nakamura. GaN growth using GaN buffer layer. *Japanese Journal of Applied Physics*, 30(10A):L1705, 1991.
- [146] Y. Zhong, K.S. Wong, W. Zhang, and D.C. Look. Radiative recombination and ultralong exciton photoluminescence lifetime in GaN freestanding film via two-photon excitation. *Applied Physics Letters*, 89(2):022108, 2006.
- [147] H. Mahr and M.D. Hirsch. An optical up-conversion light gate with picosecond resolution. *Optics Communications*, 13(2):96–99, 1975.
- [148] M.A. Kahlow, W.O. Jarzęba, T.P. DuBruil, and P.F. Barbara. Ultrafast emission spectroscopy in the ultraviolet by time-gated upconversion. *Review of Scientific Instruments*, 59(7):1098–1109, 1988.
- [149] J. Shah, T.C. Damen, B. Deveaud, and D. Block. Subpicosecond luminescence spectroscopy using sum frequency generation. *Applied Physics Letters*, 50(19):1307–1309, 1987.
- [150] S. Kazim, M.K. Nazeeruddin, M. Grätzel, and S. Ahmad. Perovskite as light harvester: a game changer in photovoltaics. *Angewandte Chemie International Edition*, 53(11):2812–2824, 2014.
- [151] C. de Weerd, L. Gomez, H. Zhang, W.J. Buma, G. Nedelcu, M.V. Kovalenko, and T. Gregorkiewicz. Energy transfer between inorganic perovskite nanocrystals. *The Journal of Physical Chemistry C*, 120(24):13310–13315, 2016.
- [152] P. Guyot-Sionnest, M. Shim, C. Matranga, and M. Hines. Intraband relaxation in CdSe quantum dots. *Physical Review B*, 60(4):R2181, 1999.
- [153] P. Kambhampati. Hot exciton relaxation dynamics in semiconductor quantum dots: radiationless transitions on the nanoscale. *The Journal of Physical Chemistry C*, 115(45):22089–22109, 2011.
- [154] R.A. Smith. *Semiconductors*. Cambridge Press, 1959.
- [155] K. Wu, G. Liang, Q. Shang, Y. Ren, D. Kong, and T. Lian. Ultrafast interfacial electron and hole transfer from CsPbBr_3 perovskite quantum dots. *Journal of the American Chemical Society*, 137(40):12792–12795, 2015.

- [156] M.B. Price, J. Butkus, T.C. Jellicoe, A. Sadhanala, A. Briane, J.E. Halpert, K. Broch, J.M. Hodgkiss, R.H. Friend, and F. Deschler. Hot-carrier cooling and photoinduced refractive index changes in organic-inorganic lead halide perovskites. *Nature Communications*, 6:8420, 2015.
- [157] P. Zhang, Y. Feng, X. Wen, W. Cao, R. Anthony, U. Kortshagen, G. Conibeer, and S. Huang. Generation of hot carrier population in colloidal silicon quantum dots for high-efficiency photovoltaics. *Solar Energy Materials and Solar Cells*, 145:391–396, 2016.
- [158] Y. Rosenwaks, M.C. Hanna, D.H. Levi, D.M. Szymyd, R.K. Ahrenkiel, and A.J. Nozik. Hot-carrier cooling in GaAs: Quantum wells versus bulk. *Physical Review B*, 48(19):14675, 1993.
- [159] R.D. Schaller and V.I. Klimov. High efficiency carrier multiplication in PbSe nanocrystals: implications for solar energy conversion. *Physical Review Letters*, 92(18):186601, 2004.
- [160] N.S. Makarov, S. Guo, O. Isaienko, W. Liu, I. Robel, and V.I. Klimov. Spectral and dynamical properties of single excitons, biexcitons, and trions in cesium–lead-halide perovskite quantum dots. *Nano Letters*, 16(4):2349–2362, 2016.
- [161] A.V. Barzykin and M. Tachiya. Stochastic models of charge carrier dynamics in semiconducting nanosystems. *Journal of Physics: Condensed Matter*, 19(6):065105, 2007.
- [162] Y. Wang, X. Li, J. Song, L. Xiao, H. Zeng, and H. Sun. All-inorganic colloidal perovskite quantum dots: A new class of lasing materials with favorable characteristics. *Advanced Materials*, 27(44):7101–7108, 2015.
- [163] Y.S. Park, S. Guo, N.S. Makarov, and V.I. Klimov. Room temperature single-photon emission from individual perovskite quantum dots. *ACS Nano*, 9(10):10386–10393, 2015.
- [164] L.A. Padilha, J.T. Stewart, R.L. Sandberg, W.K. Bae, W.K. Koh, J.M. Pietryga, and V.I. Klimov. Carrier multiplication in semiconductor nanocrystals: influence of size, shape, and composition. *Accounts of Chemical Research*, 46(6):1261–1269, 2013.
- [165] V.I. Klimov. Optical nonlinearities and ultrafast carrier dynamics in semiconductor nanocrystals. *The Journal of Physical Chemistry B*, 104:6112–6123, 2000.
- [166] P. Kambhampati. Unraveling the structure and dynamics of excitons in semiconductor quantum dots. *Accounts of Chemical Research*, 44(1):1–13, 2010.
- [167] E. Rabani, D.R. Reichman, P.L. Geissler, and L.E. Brus. Drying-mediated self-assembly of nanoparticles. *Nature*, 426(6964):271–274, 2003.
- [168] S. Yakunin, L. Protesescu, F. Krieg, M.I. Bodnarchuk, G. Nedelcu, M. Humer, G. De Luca, M. Fiebig, W. Heiss, and M.V. Kovalenko. Low-threshold amplified spontaneous emission and lasing from colloidal nanocrystals of caesium lead halide perovskites. *Nature Communications*, 6:8056, 2015.
- [169] M. Saba, S. Minniberger, F. Quochi, J. Roither, M. Marceddu, A. Gocalinska, M.V. Kovalenko, D.V. Talapin, W. Heiss, A. Mura, and G. Bongiovanni. Exciton–exciton interaction and optical gain in colloidal CdSe/CdS dot/rod nanocrystals. *Advanced Materials*, 21(48):4942–4946, 2009.
- [170] F.H. Alharbi and S. Kais. Theoretical limits of photovoltaics efficiency and possible improvements by intuitive approaches learned from photosynthesis and quantum coherence. *Renewable and Sustainable Energy Reviews*, 43:1073–1089, 2015.
- [171] M.C. Hanna and A.J. Nozik. Solar conversion efficiency of photovoltaic and photoelectrolysis cells with carrier multiplication absorbers. *Journal of Applied Physics*, 100(7):074510, 2006.
- [172] J.W. Robinson, E.M. Skelly Frame, and G.M. Frame II. *Undergraduate Instrumental Analysis*. Marcel Dekker, 2005.
- [173] A. Swarnkar, A.R. Marshall, E.M. Sanehira, B.D. Chernomordik, D.T. Moore, J.A. Christians, T. Chakrabarti, and J.M. Luther. Quantum dot–induced phase stabilization of α -CsPbI₃ perovskite for high-efficiency photovoltaics. *Science*, 354(6308):92–95, 2016.
- [174] R.J. Ellingson, M.C. Beard, J.C. Johnson, P. Yu, O.I. Micic, A.J. Nozik, A. Shabaev, and A.L. Efros. Highly efficient multiple exciton generation in colloidal PbSe and PbS quantum dots. *Nano Letters*, 5(5):865–871, 2005.
- [175] J.E. Murphy, M.C. Beard, A.G. Norman, S.P. Ahrenkiel, J.C. Johnson, P. Yu, O.I. Mićić, R.J. Ellingson, and A.J. Nozik. PbTe colloidal nanocrystals: synthesis, characterization, and multiple exciton generation. *Journal of the American Chemical Society*, 128(10):3241–3247, 2006.
- [176] S. Saeed, C. de Weerd, P. Stallinga, F.C.M. Spoor, A.J. Houtepen, L.D.A. Siebbeles, and T. Gregorkiewicz. Carrier multiplication in germanium nanocrystals. *Light: Science & Applications*, 4(2):e251, 2015.
- [177] M. Aerts, T. Bielewicz, C. Klinke, F.C. Grozema, A.J. Houtepen, J.M. Schins, and L.D.A. Siebbeles. Highly efficient carrier multiplication in PbS nanosheets. *Nature Communications*, 5:3789, 2014.

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 $\sim 1-50\text{nm}$), 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 ~ 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 $\sim 1-50\text{nm}$), 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 de belangrijkste halfgeleider in de hedendaagse maatschappij, besproken als een mogelijke oplossing om de fundamentele fotonvoltaïsche omzettingsefficiëntielimiet te overwinnen (de Shockley-Queisser limiet). Grote verliezen vinden plaats aan beide kanten van het zonnenspectrum 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 fotonvoltaïsche 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 ~ 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 SiO₂ 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 Er³⁺ ionen te onderzoeken. De nadruk ligt vooral op de snelle energieoverdracht van de Si nanokristallen naar de laagst aangeslagen energietoestand van het Er³⁺ 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 onderzoeksobject 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

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 SiO₂ 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 Er³⁺ 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 zonnecel-efficiëntie 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 SiO₂ 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 bio-functional upconversion nanoplatforms*, The Journal of Physical Chemistry Letters 6(13):2518, 2015.
7. E.M.L.D. de Jong, S. Saeed, W.C. Sinke and T. Gregorkiewicz. *Generation of hot carriers for photon management in future photovoltaics*, Solar Energy Materials and Solar Cells 135:67, 2015 - Chapter 2.
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. *Multielectron lifetime in all-inorganic CsPbBr₃ 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 SiO₂ 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.

12. E.M.L.D. de Jong, G. Mannino, T. Gregorkiewicz and G. Faraci. *Effect of phosphorous doping on the optical properties of large faceted Si crystalline nanoparticles (working title)*, in preparation - Chapter 5.
13. E.M.L.D. de Jong et al. *Energy transfer dynamics in SiO₂:Er sensitized with Si nanocrystals (working title)*, in preparation - Chapter 6.
14. E.M.L.D. de Jong, G. Yamashita, J. Takatsu, M. Ashida, Y. Fujiwara and T. Gregorkiewicz. *Investigations of the free-carrier dynamics in Eu-doped GaN grown by OMVPE (working title)*, in preparation - Chapter 7.
15. E.M.L.D. de Jong, G. Yamashita, L. Gomez, M. Ashida, Y. Fujiwara and T. Gregorkiewicz. *Ultra-fast optical spectroscopy of carrier dynamics and multiplication in all-inorganic CsPbX₃ perovskite nanocrystals (working title)*, in preparation - Chapter 8.
16. E.M.L.D. de Jong, I. Camps, R. Serna and T. Gregorkiewicz. *Optical investigations of Eu-doped thin films based on SiAlON and Al₂O₃ (working title)*, in preparation.

Acknowledgements

The work described in this dissertation is certainly a collaborative enterprise; it has been completed due to the time, effort and support of many people, in science and outside academia. I owe deep thanks to many people for their contribution to this thesis and scientific papers, and, possibly more important, to my personal and professional development. Although I cannot thank everybody enough for what they have done for and meant to me, I will try to acknowledge them on these last two pages of my thesis.

First and foremost, I would like to express my deepest gratitude to Prof. Tom Gregorkiewicz. I feel very fortunate to have you as my supervisor and promotor. Throughout my doctoral studies, I learned a lot from you on a scientific as well as a personal level, and I will always be grateful for that. Your door is always open, and I cannot think of a moment you were unwilling to help. Thanks for all the opportunities you have given me.

I will be forever thankful to all current and former members of “Tom Gregorkiewicz’s Group” (TGG, including the bachelor and master students). In my opinion, TGG stands also for “The Greatest Group”. Thank you all for your enormous support and the collegial atmosphere! Special thanks goes to my dear paranymphs, Arnon and Bart, for their support and for accepting my invitation to stand next to me during my defense.

During my PhD studies, I have been fortunate to collaborate with many excellent researchers over the world. I have been very lucky to be able to perform measurements at Osaka University. Professor Fujiwara, Prof. Ashida and all the members of these two research groups, thank you very much for having me in your group, helping me in your lab, and introducing me to the rich Japanese culture. It has been an experience I will never forget. Arigatō gozaimashita. I am grateful to my collaborators from St. Petersburg (Prof. Yassievich, Dr. Poddubny, Dr. Gert and Dr. Prokofiev) for their invaluable input with theoretical models, which helped us to understand the experimental data and to improve our scientific papers. Moreover, I would like to thank Prof. Valenta from Prague for initiating and contributing to the interesting project on the excitation power dependence of the photoluminescence intensity of silicon nanocrystals. Additionally, I wish to acknowledge my collaborators from Catania, especially Prof. Faraci and Dr. Mannino, for their collaboration, which started when we learned about their interesting samples at the EMRS conference in Lille. I also wish to thank the group of Prof. Serna from Madrid for providing their samples and for their collaboration, and the group of Prof. Fujii from Kobe for sharing their sample expertise.

Also in the Netherlands I have been lucky to perform measurements at and collaborate with scientists in other departments and universities. I would like to thank the optoelectronic materials group of Prof. Siebbeles and the photovoltaic materials and devices group of Prof. Zeman at the TU Delft. In addition, I wish to thank two groups of the VU in Amsterdam, namely the group of Dr. von Hauff and the group of Prof. Kennis for discussions on perovskites and their support during my induced absorption measurements in their lab, respectively. Furthermore, I would like to acknowledge the molecular photonics research group (including the technicians Michiel Hilbers and Paul Reinders) at the HIMS department of the UvA for the collaborations. Several researchers have come to our lab to perform measurements and, vice versa, they have helped us with important measurements in their labs.

I am very appreciative of the financial support I received through Dutch technology foundation STW, which is part of the Netherlands organisation for scientific research (NWO) and which is partly funded by the Ministry of Economic Affairs, and the international joint research promotion program of Osaka University. I would like to thank the STW User Committee members for their valuable input during the half-yearly meetings, and for their time and effort they invested in our projects. Moreover, I wish to thank my promotion committee for their willingness to serve on my PhD committee, and their time and

effort they have spent on the evaluation of my thesis.

Furthermore, I would like to say thanks to all the group leaders, staff members, technicians, and students at IoP/WZI. Deep appreciation goes also to everyone in the secretary and administrative department for their help with paper work, and the electronic and mechanical workshop for their highly valuable technical support during our measurements and with computer problems we encountered, and for showing interest in our work.

Additionally, I also would like to express my admiration and gratitude to the Bearcats (University of Cincinnati). Teachers, coaches, classmates, friends, teammates and their parents thank you very much for giving me an unforgettable college experience. Even though it is already a couple years ago, those memories and lessons are still cherished in my heart. Special thanks goes to Monty for giving me this opportunity!

Besides sitting in a dark lab, I have spent many hours in and around several swimming pools. Thanks to all (current, but also former) teammates and coaches, I have had the privilege to have fun, to swim and to work with over the years. Coaches and volunteers, thanks for all the time and effort you put in to help us, swimmers, reach our goals. It has a lasting effect on me outside the pool. I would also like to thank my friends outside academia and the swimming pool.

And then last, but certainly not least, I would like to thank my family. In particular, my three sisters, brother-in-law Jeroen, two nephews Thijmen and Wessel, and my parents. Marein, Vanya and Gerdien thank you very much for being great sisters, for giving me valuable advice and for being an example for me. En natuurlijk mijn ouders Maria en Reinier. Door jullie grote bijdrage en steun heb ik mijn tijdsintensieve sportcarrière met studeren en later promoveren kunnen combineren. Papa en mama, ik kan het niet verwoorden. Heel erg bedankt voor alles wat jullie tot nu toe voor mij hebben gedaan!

Elinore