Cutting and emitting photons with Si nanocrystals

W.D.A.M. de Boer
Van der Waals—Zeeman Institute, University of Amsterdam,
65 Valckenierstraat, NL-1018 XE Amsterdam, The Netherlands

Si nanocrystals (SiNCs) embedded in a SiO_2-matrix are intensively studied as a possible basis for photovoltaic and photonic devices, due to stability and relatively easy manufacturing. However, electron and hole relaxation and recombination processes in SiNCs are still not fully understood.

In this presentation, I will discuss two specific results recently obtained by optical spectroscopy on solid state dispersions of SiNCs in a SiO_2-matrix. The first part concerns the (absolute) quantum efficiency (i.e. number of emitted photons per absorbed photon) of photoluminescence (PL). We demonsrate that its value increases above a certain threshold value of excitation photon energy, due to a specific carrier multiplication (CM) process, termed SSQC – "space-separated quantum cutting" [1,2]. As a consequence, the theoretical conversion limit of present day first-generation photovoltaics (Shockley-Queisser limit) can be overcome. As opposed to other NC materials in which CM also has been observed, we found that the CM occurs for photon energies within the range of the solar spectrum, rendering it highly suitable for PV applications.

The second part of the presentation considers the visible emission sometimes reported for SiNCs. In the investigated materials, it appears next to the well-known excitonic emission (in the near-infrared) and the oxygen/defect-related band (at ~420 nm). Upon NC size decrease, its maximum intensity shifts towards longer wavelengths, i.e. shows a behavior opposite to the well-known "blue" shift of the excitonic emission. The "red" spectral shift for smaller nanocrystal sizes is accompanied by increase of intensity, indicating enhancement of quantum efficiency. Based on the detailed experimental study, we identify the newly observed band – the "hot" emssion – with radiative no-phonon recombinations of non-equilibrium electron-hole pairs, whose properties are influenced by quantum confinement [3]. We also show that the efficiency of this hot emission band is enhanced by 3 orders of magnitude when compared to bulk Si. We discuss the most important consequences of this finding for understanding of quantum confinement effects in Si nanocrystals, and its application impact.

1. D. Timmerman *et al.*, Nature Photonics 2, 105 - 109 (2008)
2. D. Timmerman *et al.*, Nature Photonics, *under submission*
3. W.D.A.M de Boer *et al.*, Nature Nanotechnology, *accepted for publication*