Energy relaxation in optically excited Si and Ge nanocrystals
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CHAPTER 9

Outlook
The scientific objective of the research presented in this thesis is to explore energy relaxation processes of optically excited Si and Ge nanocrystals. A fundamental understanding of these phenomena is an essential condition before these materials can be used for practical applications. The identification and deeper understanding of unique energy relaxation paths obtained in this study will open a new window of opportunity for these materials. In particular, in this work a different way of harvesting energy, which is typically lost to heat in conventional photovoltaic devices, is proposed. These losses are mainly due to inefficient conversion of photons in the ‘blue’ part of the solar spectrum. Our results demonstrate a method to transform the high-energy photons into multiple smaller ones, whose photovoltaic conversion can be more efficient. The present schemes of hot-carrier solar cells propose to use direct extraction of ‘hot’ charges created upon absorption of high-energy photons through resonant contacts. This is extremely challenging, since the extraction of hot carriers has to compete with their ultrafast thermalization. Here, an attractive alternative conversion of the excess energy into infrared photons is proposed. It is demonstrated that this ‘optical cooling’ can successfully compete with heat generation. As a result, high-energy photons are streamlined into infrared quanta whose energy can be optimized for the highest conversion efficiency. The proposed scheme makes use of Si nanocrystals and the popular rare-earth element erbium, thoroughly investigated in the past for possible uses in telecommunication networks. The choice follows from theoretical evaluations based on similar consideration for the multiple exciton generation and predicts the highest efficiency upon spectral conversion into 0.8 eV quanta — an energy equal to that of the first excited state of Er$^{3+}$ ion. Our findings form the physical basis for a novel hot-carrier photovoltaic architecture which would then comprise (i) a standard Si solar cell with (ii) a non-contacted spectrum shaper in front, converting high-energy photons into IR, and (iii) a low bandgap cell on the back side for efficient harvesting of the IR photons, and would be capable of exceeding the Shockley-Queisser limit. In our investigations, we found that the external quantum yield of the IR photon generation by Er$^{3+}$ ions shows ~15-fold enhancement in the high excitation energy range. This increase is entirely due to the efficient transfer of excess energy of hot carriers. We also showed that this process can be tuned by material parameters — the Si NC size and absolute and relative concentrations of Er$^{3+}$ ions and NCs. Another important result of this thesis is the report on the observation of carrier multiplication in Ge NCs. While carrier multiplication has been reported for NCs of many semiconductors, Ge was missing from that list. Ge features a number of unique properties, such as superior level of chemical purity, a very special and strain-tunable band structure with only a small difference between energies of
the indirect and direct bandgaps, large absorption cross-section and a large Bohr radius, among others. These prominent features make Ge interesting for photonic and electronic applications. Ge NCs also are very interesting for detectors and photovoltaics. In the latter case, if the idea of a simple single p-n junction is abandoned and devices making use of carrier multiplication are developed, Ge might become the material of choice, since Ge NCs offer the possibility for realization of the most convenient bandgap, according to theory. Our study shows that carrier multiplication:

1. does take place in Ge NCs,
2. is considerably more efficient than impact excitation in bulk Ge,
3. occurs with only a minimal energy loss.

The results obtained in the studies presented in this thesis open new directions of research and opportunities, where the most important one is the material optimization. While the processes have been demonstrated experimentally, they are not been currently applied to practical devices. The follow-up studies will focus on further improvements by tweaking material properties. In particular, this concerns increasing the conversion efficiency of photovoltaics by integrating these materials in photovoltaics.