UNIVERSITY OF AMSTERDAM

Energy Relaxation in Optically Excited Si and Ge Nanocrystals

S. Saeed

Energy relaxation in optically excited Si and Ge nanocrystals

Summary

This thesis entitled “Energy relaxation in optically excited Si and Ge nanocrystals” presents the PhD research carried out at the Van der Waals-Zeeman Institute at the University of Amsterdam. Optical spectroscopy of semiconductor nanocrystals has been conducted in order to gain insights on the energy relaxation processes going on in these quantum confined materials. This will help in development of new concepts and materials that could find their way to various applications and, e.g., increase the efficiency of photovoltaic conversion.

The first chapter starts by recalling the importance of semiconductors, in particular silicon and germanium. Quantum confinement effects in the nanometer sized silicon and germanium are discussed. These effects make nanocrystals and nanoclusters of these materials interesting for electronic and photovoltaic applications. Next, keeping an eye on some unique properties of germanium, the ‘mystery’ surrounding emission from germanium nanocrystals is briefly summarized. Rare earth doped semiconductors are discussed, especially the sensitization of erbium photoluminescence in silicon dioxide by silicon nanocrystals. Next, carrier multiplication in nanocrystals and its importance for photovoltaics is described. The first chapter ends with a short description concerning the structure of this thesis.

Chapter 2 discusses the details of the sample preparation techniques and the samples used in this study. Various instruments used for optical spectroscopy, including excitation and detection systems used for different wavelength ranges, are described. Experimental techniques for determining absorption (linear and induced) are discussed. Also the instrumentation and the scaling procedure use for obtaining the quantum yield are described.

In Chapter 3, the study of temperature-dependent photoluminescence from erbium-doped silicon dioxide sensitized with silicon nanocrystals is presented. Special emphasis is on the observation of fast erbium-related photoluminescence at 1.5 μm. This emission has been observed in the past and was assigned to the defects in SiO2 matrix. In this study, on the basis of time-resolved and temperature-dependent measurements, it is confirmed that the major part of this emission is erbium-related.

Chapter 4 presents the study of the external quantum yield of erbium-related photoluminescence from erbium-doped silicon dioxide sensitized with silicon nanocrystals. The quantum yield of this emission is constant at low pump energies and increases in a step-like manner above a certain threshold of excitation energy. The enhancement of the external quantum yield is assigned to the increased contribution of hot-carrier mediated excitation of erbium ions. In this way the excess energy of hot carriers generated in nanocrystals, otherwise lost as heat, can be optically extracted by exciting (multiple) erbium ions. Influence of the nanocrystal size and concentration, and the erbium-to-nanocrystal concentration ratio on external quantum yield of erbium-related emission is investigated

in detail and discussed. Multiple erbium ion excitation routes and the possible application scheme for photovoltaics are considered.

In Chapter 5, observation of carrier multiplication in germanium nanocrystals is described. Transient induced absorption spectroscopy is used to determine the efficiency of this process. The procedure used to establish this effect is described in detail. A Carrier multiplication rate of approximately 190% is observed at 2.8 times the optical bandgap of germanium nanocrystals. On the basis of this result, it is concluded that the carrier multiplication in germanium nanocrystal is much more efficient then the impact excitation in bulk germanium.

Chapter 6 deals with the structural and optical study of solid-state dispersions of germanium nanocrystals prepared by plasma-enhanced chemical vapor deposition. Based on the material characteristics, a possible microscopic origin of the individual emission bands is discussed and excitonic emission from very small germanium nanocrystals is suggested to be responsible for one of them. Other emission bands possibly related to defects are also identified.

Chapter 7 and 8 summarizes the results of the ongoing and still not finalized investigations by transient induced absorption spectroscopy. In Chapter 7, transient induced absorption spectroscopy of geranium nanocrystals is presented. Ultrafast carrier dynamics show interesting features, especially the observation of a prominent fast, of the order of few picosecond, negative signal in infrared and visible range of probe energies. Mechanisms that could give rise to such an effect are considered and the possible existence of additional states inside the bandgap of germanium nanocrystals is postulated.

Transient induced absorption spectroscopy of erbium-doped silicon dioxide sensitized with silicon nanocrystals is presented in Chapter 8. Again here, the concern is the observation of a fast negative induced absorption signal for the visible probing range. Possible explanations are postulated, while clearly further investigations are required.

Finally, an outlook of the research is given in Chapter 9.