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Exciton dynamics of silicon quantum dots –Femtosecond near-IR transient absorption spectroscopic study–

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Introduction

Silicon is an important material for application in electronic and photovoltaic industry. Recently, silicon nanoparticles attract a great deal of attention due to their size-tunable, non-toxic and electronic properties. Especially, the fascination caused by the process called multiple exciton generation (MEG) that multiple pairs of excitons could be generated just by a single high-energy photon is getting more focused by the researchers. MEG in silicon quantum dots (Si QDs) has a strong potential for application in photovoltaic devices. As a result, multiple excitons decay dominantly via Auger recombination that is a process in which an exciton transfers its recombination energy to another particle (an exciton, an electron or a hole) and excites it to a higher energy state. Auger recombination rate is enhanced by the relaxation of the translational momentum conservation in nanocrystals (NCs) that must to be satisfied in bulk materials [1]. The enhancement of Auger recombination rate in NCs diminishes the potential of MEG in photovoltaics as well as for light sources [2] as it reduces the lifetime of multiple excitons localized in the same NC. So it is very important to reduce or suppress Auger recombination rate in NC materials. For that purpose, microscopic understanding of multibody Auger recombination in quantum confined system is important both from fundamental viewpoints and for applications. In the present study, we prepared several kinds of silicon nanoparticles by a ball milling method, and exciton dynamics and Auger recombination of silicon nanoparticles were examined by femtosecond near-IR transient absorption spectroscopy.generated

Experiments

We prepared three Si NCs synthesized via a ball milling method, and dissolved in the ethanol (80%) + water (20%), propanol, and propanol (80%) + water (20%), respectively. Each samples were tagged as Si A, Si B and Si C. Transmission electron microscopy (TEM) was used to analyze the size of Si NCs. The samples were excited at second harmonic (400 nm) of an amplified Ti: sapphire laser (800 nm, 60 fs and 500 Hz) for femtosecond near-IR transient absorption spectroscopy. All the experiments were conducted at room temperature.

Results and Discussion

To estimate the bandgap energy (Eg) of Si NCs, absorption spectra of each samples were transferred into the graph of $(h\nu\alpha)^{1/2}$ against photon energy as similar to the method mentioned Meier's Group [3], where α is an absorption coefficient. The results were illustrated in Fig. 1. Eg of Si A, Si B, and Si C were estimated to be 1.85, 1.76 and 1.70 eV,

respectively. A typical TEM image of Si A is shown in the inset of Fig. 1, where the average diameter is estimated to be 2.6 nm. This value is little smaller than the size estimated from Eg (3.1nm) [4]. The excitation energy at 400 nm (3.1 eV) is below the MEG threshold (<2 Eg), so the multiple excitons in one Si NC could appear only by absorption of multiple photons. The transient absorption spectra of Si NCs are very broad and positive signal in near-IR region (900 ~ 1400 nm), suggesting the intraband transition in the conduction band. Fig.2 illustrates the transient absorption dynamics of Si C sample at various excitation intensities observed at 1000 nm. The dynamics were globally analyzed by two or three exponential decay functions. The fast decay component of 0.5 ~ 0.8 ps was detected as a main component irrespective of a kind of Si NCs (A ~ C) and excitation intensity. The fast component is probably due to the carrier trapping by the defects. Slow component of a few tens ps was also observed. However, the amplitude of a fast component increased with increasing the excitation intensity. This result suggests that multiple excitons are easily trapped to the defect sites in Si NCs.

References

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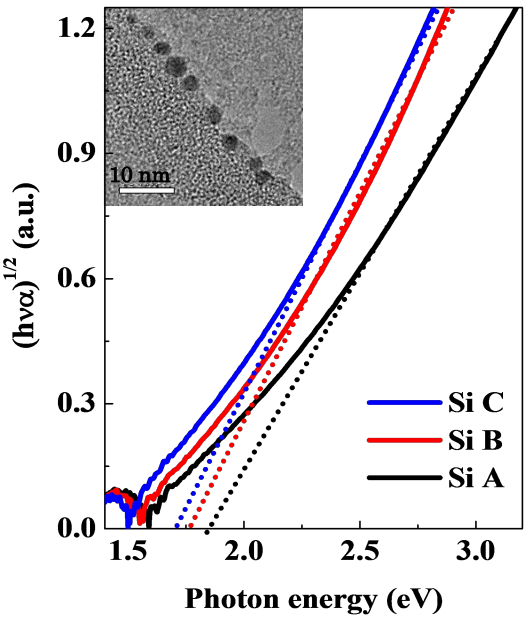

Fig. 1 $(h\nu\alpha)^{1/2}$ of Si A, Si B and Si C are plotted as a function of photon energy. TEM image of Si A is shown in the inset.

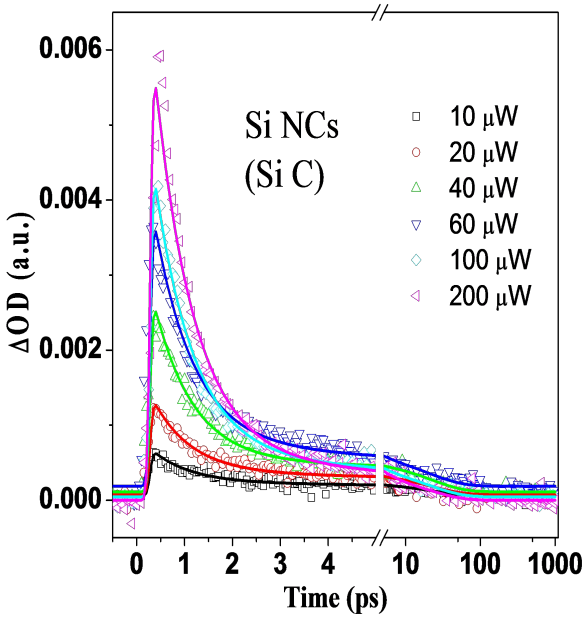

Fig. 2 The transient absorption dynamics of Si C at different excitation intensities observed at 1000 nm.