

# 博士論文（要約）

Exciton Diffusion and Photon Antibunching in Carbon Nanotubes  
(カーボンナノチューブにおける励起子拡散と光アンチバンチング)

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## Abstract

Single-walled carbon nanotubes are emitters at telecom wavelengths which can be directly grown on silicon, being promising materials for nanoscale optoelectronic devices. Due to limited screening of Coulomb interaction in nanotubes, excitons play a key role in light emission, and therefore understanding the excitonic properties is important.

In particular, luminescence properties of carbon nanotubes are strongly affected by exciton diffusion, which plays an important role in various nonradiative decay processes. Here we perform photoluminescence microscopy on hundreds of individual air-suspended carbon nanotubes to elucidate the interplay between exciton diffusion, end quenching, and exciton-exciton annihilation processes. A model derived from random-walk theory as well as Monte Carlo simulations are utilized to analyze nanotube length dependence and excitation power dependence of emission intensity. We have obtained the values of exciton diffusion length and absorption cross section for different chiralities, and diameter-dependent photoluminescence quantum yield have been observed. The simulations have also revealed the nature of a one-dimensional coalescence process, and an analytical expression for the power dependence of emission intensity is given.

In addition, carbon nanotubes have great potential for single photon sources as they have stable exciton states even at room temperature and their emission wavelengths cover the telecommunication bands. In recent years, single photon emission from carbon nanotubes has been achieved by creating localized states of excitons. In contrast to such an approach, here we utilize mobile excitons and show that single photons can be generated in air-suspended carbon nanotubes, where exciton diffusion length is as long as several hundred nanometers and exciton-exciton annihilation is efficient. We perform photoluminescence microscopy on as-grown air-suspended carbon nanotubes in order to determine their chirality and suspended length. Photon correlation measurements are performed on nanotube emission at room temperature using a Hanbury-Brown-Twiss setup with InGaAs/InP single photon detectors. We observe antibunching with a clear excitation power dependence, where we obtain  $g^2(0)$  value less than 0.5 at low excitation powers, indicating single photon generation. We show such  $g^2(0)$  data with different chiralities and suspended lengths, and the effects of exciton diffusion on single photon generation processes are discussed.

Finally, we explore a possibility of integration of carbon nanotube dopant emitters with silicon microcavities, which is another promising pathway to carbon nanotube applications as single photon emitters. We fabricate two-dimensional photonic crystal microcavities on silicon-on-insulator substrates with an appropriate cavity mode design for dopant state emission of (6,5) carbon nanotubes. Doped carbon nanotubes are deposited on the cavities using a micromanipulation system, and photoluminescence enhancement of the dopant states is observed by optical spectroscopy. We perform lifetime measurements using a pulsed laser and superconducting nanowire single photon detectors for devices with good optical coupling, and the contribution of cavity resonance is evaluated.