In-situ Observation of Raman Spectrum of Isolated Single-Wall Carbon Nanotube on Au Electrode

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Introduction

Since the discovery of carbon nanotubes, a great deal of activities has been focused on this new nanoscale material. In particular, determination of the electronic structures of individual single-walled carbon nanotubes (SWNTs) is a central issue. Depending on the geometrical structures and their chiralities of the tubes, the electronic structures change drastically to be metallic or semiconducting. Raman spectroscopy is one of the powerful techniques to characterize SWNT, because each SWNT shows specific Raman spectrum reflecting their electronic structure. [1] These spectra are realized in terms of resonance enhancement and optical band-gap excitation between van Hove singularities in the electronic density of states. In the present study, we used in-situ resonant confocal micro Raman spectroscopy to determine the potential dependent change in the Raman intensity of isolated SWNT on Au electrode under electrochemical potential control in aqueous solution.

Experimental

Adsorption of isolated SWNT onto a cleaned polycrystalline Au surface was carried out by dropped highly dispersed SWNT solution containing sodium dodecyl sulfate (SDS). [2] Confocal resonance near-infrared (NIR) Raman measurements carried out using 785 nm (Eex = 1.58 eV) excitation with the output intensity of 1mW. Estimated spot size of the irradiation was approxymately1 μm. SWNT adsorbed Au electrode was fixed at the bottom of a Teflon cell, which filled deaerated aqueous 0.2 M Na_2SO_4 electrolyte (Figure 1). A platinum wire was set as a counter electrode. The potential of the Au working electrode was regulated with respect to the Ag/AgCl reference electrode. The potential was swept with the rate of 10 mV/s in the range between –800 mV and +400 mV.

Result and Discussion

Atomic force microscope measurement showed that the present sample have a concentration of < 1.0 SWNT / μm^2 with tube lengths from 200 nm to 2000 nm on the Au substrate. Figure 2 shows that typical Raman spectrum of isolated single SWNT in the electrolyte solution. Observation of the sample at each spot gave typical Raman spectrum of isolated single SWNT, showing radial breathing mode (RBM) in the low frequency region around 100-300 cm^{-1}, and D-, G-, and G'- bands in 1200-2600 cm^{-1}. [3] Spectra at different spots gave different peaks of RBM. The full width at half maximum (FWHM) at the RBM peaks of metallic tube around 140-180 cm^{-1} and semiconducting tube around 200-240 cm^{-1} were approximately 9 cm^{-1} and 3 cm^{-1}, respectively. These characteristics suggest that the observed spectrum at each spot reflect resonance NIR Raman scattering intensity of individual SWNT on the Au electrode. Apparent feature of asymmetric profile towards lower frequency at G-band, a Breit-Wigner-Fano (BWF) profile, around 1580 cm^{-1} was not observed at most of the spots, indicating that contribution of bundled tubes and electron-phonon interaction between SWNT and Au electrode should be minor in the present system. The sample of the highly dispersed SWNT on the Au electrode allow us to observe the characteristics of the electronic states of isolated individual SWNT whose charge densities are controlled by the electrochemical potential of the Au electrode.

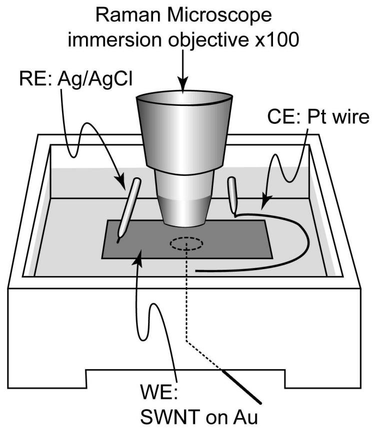

Figure 1. Electrochemical cell.

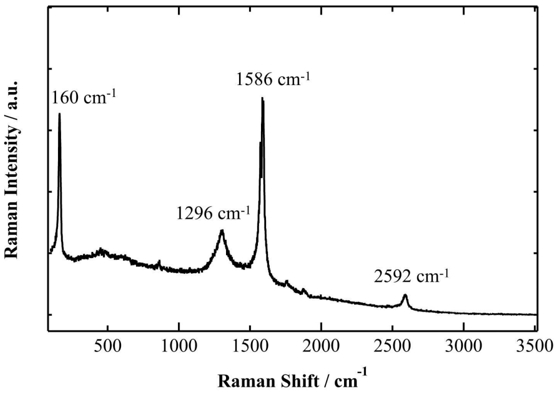

Figure 2. Raman spectrum of isolated SWNT adsorbed on Au electrode in aqueous 0.2 M Na_2SO_4 solution; Electrode potential was kept at 0 mV vs. Ag/AgCl.

Reference

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