Anomalous resonant Raman scattering from vertically aligned SWNT films

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Recently, we have developed a method to synthesize vertically aligned SWNT film with centimeter-order uniformity [1]. Since the film has several μ m thickness, it is quite possible to perform polarized Raman analysis with sufficient signal intensity (Fig. 1). With this unique specimen, we demonstrate the shape of RBM spectrum exhibits a strong dependence on the polarization over SWNT axis (Fig. 2). Moreover, RBM peaks commonly observed with 488, 514.5, 633 nm lasers are found to be classified into two types according to the polarization dependence. Since the laser input was chosen sufficiently low (250 ~ 2500 W/cm⁻², more than

3 order lower than typical micro-Raman experiment [2]), this change is not due to the heating of SWNTs. This strong polarization dependence of RBM could be explained by a non-vertical $(\mu \rightarrow \mu \pm 1)$ excitation caused by absorption of cross-polarized light recently discussed by Grüneis *et al.* [3], who observed peaks apparently deviated from the Kataura plot at lower energy region from their randomly oriented SWNT ropes [4]. The spectral change were analyzed and made it possible to unambiguously classify the types of RBM peaks even at higher energies (1.96 - 2.54 eV). The grouping behavior of RBM peaks was supported by a molecular adsorption experiment that can cause similar spectral change within the "from top" configuration.

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- [2] A. Jorio et al., New J. Phys. 5 (2003) 139.
- [3] A. Grüneis et al., Phys. Rev. B 67 (2003) 165402.
- [4] A. Grüneis et al., Chem. Phys. Lett. 387 (2004) 301.

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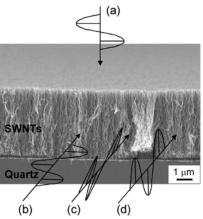


Fig. 1. Cross-sectional SEM image of vertically aligned SWNT film grown on a quartz substrate. The SWNTs form bundles with typical diameter of 15 nm. (a) \sim (d) denotes polarization and incident direction of laser light used in Fig. 2.

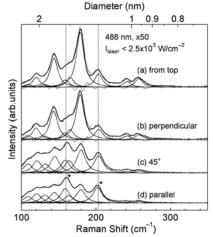


Fig. 2. Polarization dependence of RBM spectra taken with 488 nm. Each spectrum was decomposed into Lorentzian keeping FWHM the same. Intensities were normalized by G-band.