Polarized Raman and Photoluminescence Characterization of Vertically Aligned Single-Walled Carbon Nanotubes

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Polarized Raman and photoluminescence studies were employed to characterize vertically aligned single-walled carbon nanotubes (VA-SWNTs) synthesized by the alcohol catalytic CVD (ACCVD) method [1,2]. The high-purity VA-SWNT film with average nanotube diameter of 2 nm can be grown up to 30 μm with in-situ observation of film thickness [3]. When the film is excited from the top surface, we found several strong RBM peaks, i.e. a peak at 180 cm^{-1} for 488 nm excitation, are distinguishing feature of VA-SWNTs. In our previous polarized Raman studies, we found these peaks are strong for incident light polarized perpendicular to the tube growth direction but diminishes when incident light is polarized parallel to the tube growth direction [4]. We concluded that we are observing cross-polarized excitation. However, recent higher-resolution Raman measurements reveal that some of these peaks are composed of several separate fine peaks. Also, this strong peak disappears when the VA-SWNT film is dispersed. With our recent findings that our film is composed of small bundles [5], typically 5-8 nanotubes, the distinguishing Raman feature is further examined by updated polarized Raman measurements.

Given the Z-direction to be defined as parallel to the SWNT axis (growth direction), both incident and scattered light propagate along the Y-direction. After testing Y(ZZ)Y, Y(ZX)Y, Y(XX)Y, and Y(XZ)Y configurations, it was found that the RBM peaks are much stronger in the Y(XX)Y configuration (when the incident light and scattered light are perpendicular to the tube axis) than in the Y(ZZ)Y configuration. Also, the peak intensities are suppressed dramatically for the cross-polarized case, Y(XZ)Y, which is quite different from the previously well accepted result that optical spectra are dominated by absorption/emission of light polarized parallel to the tube axis, known as the antenna effect. On the other hand, G-band is much stronger for Y(ZZ)Y configuration compared with Y(XX)Y configuration. Hence, the intensity ratio between the RBM mode and G band is also larger in the Y(XX)Y case than any other configuration. We can also identify a decomposed component of G-band which is only observed for Y(ZZ)Y configuration.

Higher order Raman signal such as 2D, 2G, G+2D, 3G, 4D, 2G+2D... are observed from this VA-SWNT film with the typical photoluminescence measurement setup using Xe lamp excitation. These higher order Raman scatterings are resonant with exciton E11 energy and overlapped to the photoluminescence signal. A VA-SWNT film made of 13C isotopes synthesized by our improved

ACCVD condition is used to clarify these overlapped features.

References:

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