Optical Activity in Single-Walled Carbon Nanotubes

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Optical activity was observed in films of vertically aligned single-walled carbon nanotubes (SWNTs). We report the control of both the polarization state and transmission of incoming light at 1550nm by azimuthal and axial tilting of SWNT film about its aligned axis. The experiments reveal that the polarization state of light is susceptible to the azimuthal angle of aligned direction of SWNT having semi-conductor characteristics and the intensity of output beam after SWNT film shows cosine function dependence on axial tilting angle.

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1. Introduction

Single-walled carbon nanotubes (SWNTs) have been the subject of focused interdisciplinary studies due to their unique electrical, mechanical and thermal properties that could lead to wide and versatile applications.[1] However in optical perspectives SWNT characterization studies, not to mention practical applications, have been limited and it is only recent that SWNT could find applications in saturable absorbers for mode locking of erbium doped silica fiber lasers.[2,3] In terms of fabrication process of SWNT, significant improvements in the tube growth direction control have been recently achieved and highly polarization dependent optical transmission characteristics were reported.[4,5] Anisotropy in thin film has been extensively applied in light polarization control and, for example, Feussner-type polarizer technologies are now well established using stretched polyethylene-terephthalate films.[6] In order to meet the requirements for environment robustness, however, organic films need to be replaced by inorganic material, especially for optical communication and sensing, and it is highly expected that anisotropic carbon nano tube films would serve the purpose.

In this paper we report the optical activity of vertically aligned SWNT film in the range of 1550nm, for the first time to the best knowledge of the authors, and polarization changes are investigated on Poincare sphere as a function of azimuthal and axial tilt angle.

2. Experimental Results

In this study, SWNTs were synthesized by the alcohol-CVD (ACCVD) technique.[7] Bimetallic Co-Mo catalyst was supported on a quartz substrate by dip-coating in an acetate solution.[4] The catalyst acetate converted into nano-scale oxide where SWNT grows in vertical direction. SEM image of the grown SWNT film is shown in Fig. 1-(a). Single wall structure was confirmed by

Raman scattering spectra in Fig. 1-(b), where prominent G-band near 1593 cm^{-1} compared with negligible D-band near 1340 cm^{-1} was observed corresponding to the in-plane oscillation of carbon atoms in the graphene wall of SWNT. The radial breathing mode (RBM) in the lower frequency range as in the inset of Fig. 1 shows that the diameters of SWNTs range from 1-3 nm.

The experimental setup for measurement of optical activity in SWNTs is shown in Fig. 2. The 1550nm light emitted from a laser diode was coupled to linear polarizer (LP) to manage the state of polarization (SOP) of the input light. The optical fibers attached in LP were kept straight to prevent any perturbation in SOP of the input light. The linearly polarized input light after LP was then focused on the quartz substrate with vertically aligned SWNTs through a short length collimator. The substrate of SWNTs was mounted on rotating stage to provide tilting in the azimuthal angle, θ, and axial angle, φ, with respect to the light propagation axis, which varies the effective interaction direction and area between polarized input light and SWNTs. Output light through SWNTs was collected by another collimator and coupled to single mode optical fiber. The output polarization state of the light is then analyzed by a polarimeter (Thorlabs, PA430) along with an optical power meter. Therefore, in our setup, detailed change of SOP for various tilting angles can be traced. The power of incident light on SWNTs was kept at 300μW, which is below the threshold for saturable absorption.

The change of SOP through SWNT was measured for various tilt angle φ and corresponding traces on the Poincare sphere are shown in Fig. 3-(a). The outermost circle in the figure shows SOP through a blank quartz substrate and shows the characteristics of LP used in our experiments.

For SWNT film, SOP indeed showed a significant change as the tilt angle, φ, varied, and the changes got more prominent as the substrate made a shallow angle with respect to the incident direction. This behavior is consistent with prior report observed at 488nm, where anisotropy in transmission increased for a shallower angle. In fact if φ is over 36°, we could not distinguish the traces of SWNT from those of blank substrate. When we place SWNTs with φ=8° between the collimators, it was found initially linearly polarized state changed to elliptically polarized state, which strongly indicate existence of optical activity through SWNT. Optical transmission also did depend on the tilt angle. The insertion loss of SWNTs at φ=8° was 14.8dB and it reduced when the tilting angle increased, for example, 5.1dB at φ=25°.

The Degree of Polarization (DOP) at each tilting angle was recorded and the maximum (MAX) and minimum (Min) of DOPs were plotted with normalized values in Fig. 3-(b). The DOP at each tilting angle varied within the range of Max. and Min. DOPs. As shown in Fig. 3-(b), the DOPs increase in a monotonous manner with decreasing tilting angle. In particular, 40% increase in Max. DOP was observed for the tilting angle of 8° compared with blank substrate. From Fig. 3 we observed that the DOP as well as SOP were in deed significantly affected by the interaction of SWNT with different tilt angles. SWNT shows semi-conductor characteristics and when vertically aligned as in Fig. 1-(a), SWNT film will produce anisotropy in the interaction with incident plane electromagnetic wave of a linear polarization.

The SOP of output light through SWNTs was also traced for various azimuthal angle, θ, see Fig. 2, in the range of $-60° < \theta < 60°$ with keeping the tilting angle at φ=10°. As shown in Fig. 4, the SOP significantly changes as a function of θ in a reproducible manner and the results manifest that the SWNTs can provide a feasible solution to control polarization of light. The transverse movement of SOP in the range of $0° < \theta < 40°$ was attributed to the incomplete alignment of SWNTs in the sample.

The intensity of output light through SWNTs showed 180° periodicity with respect to θ such as a linear polarizing controller. The normalized intensity of output light was measured by optical power meter and the results are plotted in polar and x-y plot in Fig. 5. It was found that the measured intensities showed a good agreement with $\cos^2\theta$ line in both of the plots, which has been also observed in the emission and absorption behaviors of SWNTs. [8, 9]

3. Conclusions

In summary, by tilting a vertically aligned single-walled carbon nanotube (SWNT) thin film we observed polarization change in terms of both the state of polarization (SOP) and the degree of polarization (DOP) in a reproducible manner. More prominent polarization change was obtained when SWNT made a shallower angle with respect to the incident direction. In SOP linear polarization was converted to elliptic polarization, and maximum DOP increased by more than 40% at the axial angle of 8°. Output intensity also depended on azimuthal angle in $\cos^2\theta$, consistent with typical emission of SWNT.

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Figure Captions

Figure 1. SEM image of SWNTs on quartz substrate (a) and Raman spectrum at 488nm (b).

Figure 2. Experimental setup for measuring polarization change induced by SWNT film on quartz substrate. The substrate is mounted on a rotating stage which can provide rotating and tilting SWNTs with azimuthal angle, θ, and axial angle, φ, respectively. ; LD: Laser Diode, LP: Linear Polarizer, SMF: Single Mode Fiber.

Figure 3. (a) Traces of State of Polarization (SOP) while rotating linear polarizer with varying tilting angle, φ. The traces are illustrated in inset. (b) Degree of Polarization (DOP) depending on tilting angle of substrate. The data in shaded area were obtained without SWNTs.

Figure 4. Trace of State of Polarization (SOP) while rotating SWNT film with rotating angle, $-60° < \theta < 60°$.

Figure 5. Dependence of normalized intensity of output light through SWNTs on the rotating angle, θ, in polar plot (a) and x-y plot (b). The lines in both of (a) and (b) are ideal $\cos^2\theta$ graphs and the circular symbols express the measured data.

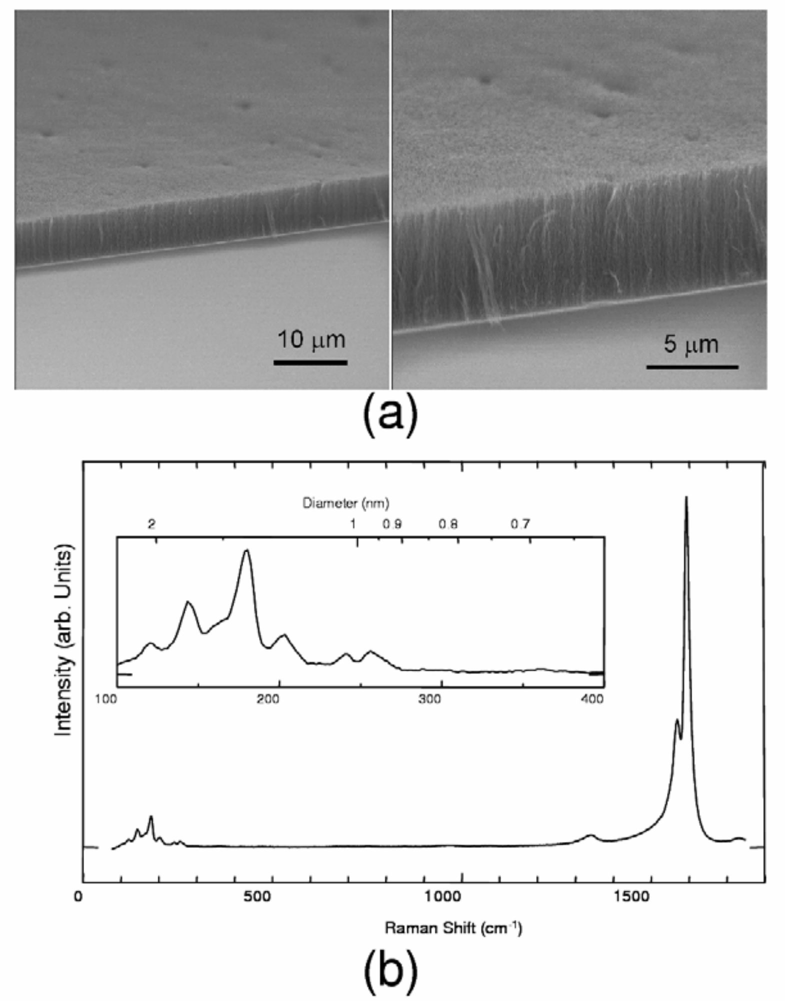
10 μm
5 μm
(a)
Diameter (nm)
2
1
0.9
0.8
0.7
100
200
300
400
Intensity (arb. Units)
0
500
1000
1500
Raman Shift (cm⁻¹)
(b)

Figure 1.

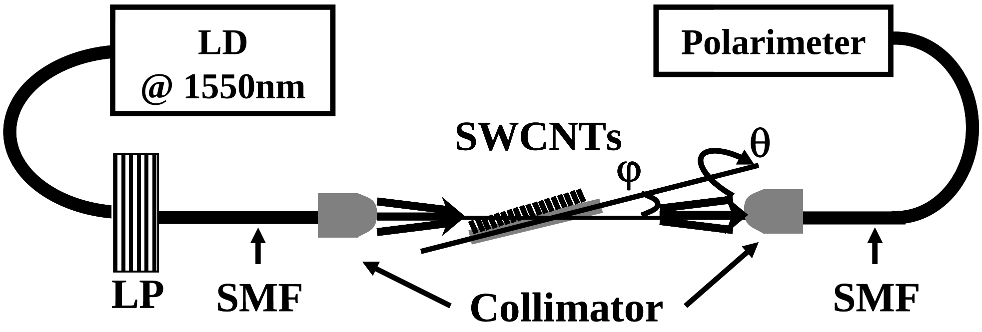

Figure 2.

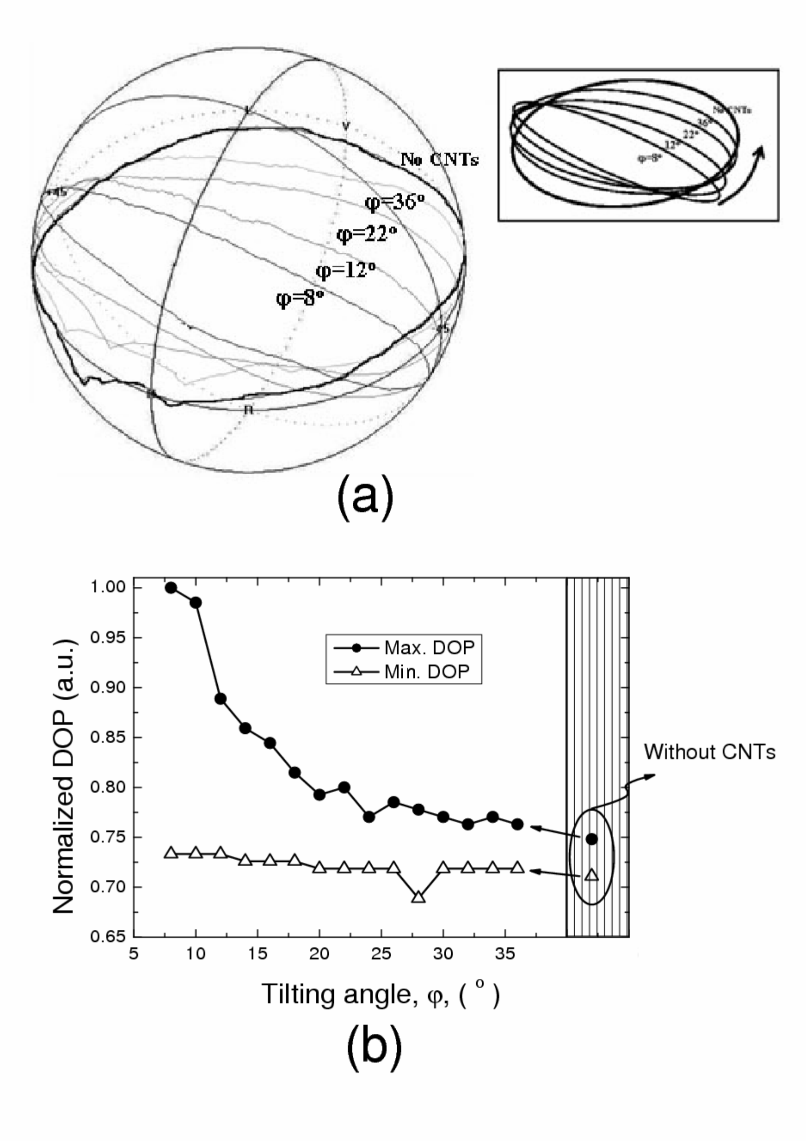
No CNTs
φ=36°
φ=22°
φ=12°
φ=8°
(a)
Max. DOP
Min. DOP
Without CNTs
Normalized DOP (a.u.)
Tilting angle, φ, (°)
(b)

Figure 3.

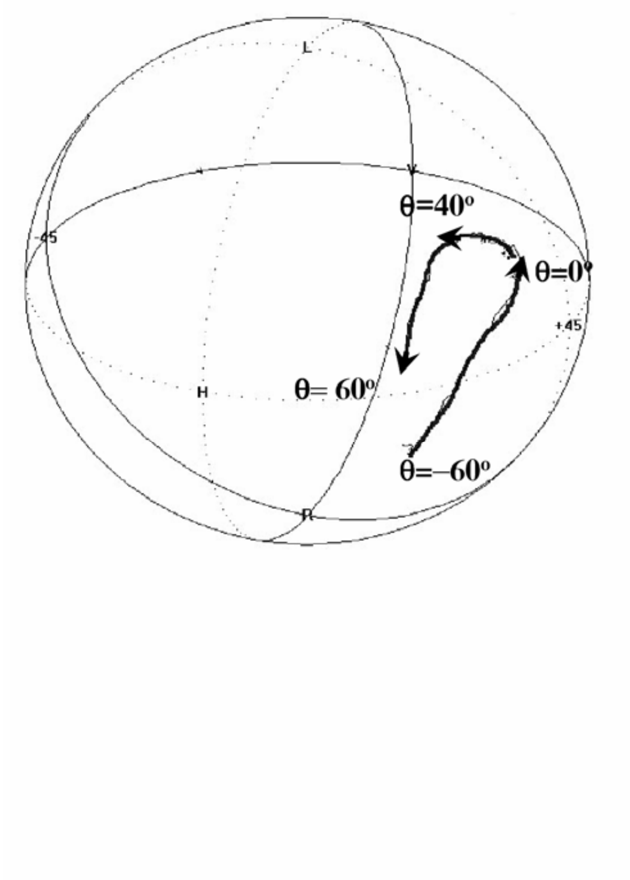

Figure 4.

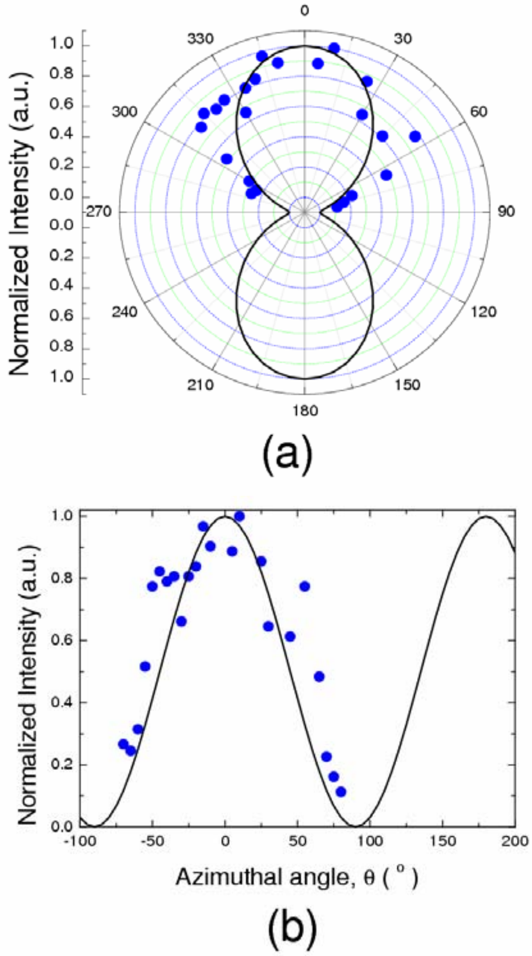
Normalized Intensity (a.u.)
0
30
60
90
120
150
180
210
240
270
300
330
1.0
0.8
0.6
0.4
0.2
0.0
(a)
Normalized Intensity (a.u.)
Azimuthal angle, θ (°)
-100
-50
0
50
100
150
200
(b)

Figure 5.