Optical anisotropy in single-walled carbon nanotubes

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Optical anisotropy at optical communication wavelength 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 1550 nm 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 the aligned direction of a SWNT having semiconductor characteristics and the intensity of the output beam after SWNT film shows cosine function dependence on the axial tilting angle. © 2005 Optical Society of America

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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 the 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 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 nanotube films would serve the purpose.

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

In this study, SWNTs were synthesized by the alcohol-CVD (ACCVD) technique.7 The bimetallic Co–Mo catalyst was supported on a quartz substrate by dip coating in an acetate solution. The catalyst acetate converted into nanoscale oxide where SWNT grows in a vertical direction. An SEM image of the grown SWNT film is shown in Fig. 1(a). A closer observation of the picture confirmed the single-wall structure. The single-wall structure was confirmed by Raman scattering spectra by 488 nm laser excitation in Fig. 1(b), where a prominent G band near 1592 cm$^{-1}$ compared with a negligible D band near 1340 cm$^{-1}$ attributed to defects or dangling bonds was observed corresponding to the in-plane oscillation of carbon atoms in the graphene wall of SWNT.4 Raman shift at 1592 cm$^{-1}$ is a unique response from SWNTs by 488 nm excitation. The radial breathing mode (RBM) in the lower-frequency range as shown in the inset of Fig. 1 shows that the diameters of SWNTs range from 1 to 3 nm.

The experimental setup for measurement of optical anisotropy in SWNTs is shown in Fig. 2. The 1550 nm light emitted from a laser diode was coupled to a fiber...
polarization rotator (FPR, 0Z Optics, FPR-11 series) to manage the state of polarization (SOP) of the input light. The polarizer is designed for 1550 nm and provides more than 30 dB extinction ratio. The optical fibers attached in the FPR were kept straight to prevent any perturbation in the SOP of the input light.

The linearly polarized input light after the FPR was then focused on the quartz substrate with vertically aligned SWNTs through a short length collimator. The substrate of SWNTs was mounted on a rotating stage to provide tilting in the azimuthal angle, \( \theta \), and axial angle, \( \phi \), 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 a single-mode optical fiber. The output polarization state of the light is then analyzed by a polarimeter (Thorlabs, PA430) along with an optical powermeter. Therefore, in our setup, a detailed change of the SOP for various tilting angles can be traced. The power of incident light on SWNTs was kept at 300 \( \mu \)W, which is below the threshold for saturable absorption.

The change of SOP through SWNT was measured for various tilting angles, \( \phi \), and corresponding traces on the Poincaré sphere are shown in Fig. 3(a). The outermost circle in the figure shows the SOP through a blank quartz substrate and shows the characteristics of the FPR used in our experiments.

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

Moreover, we characterize the degree of polarization (DOP) of the given SWNT as shown in Fig. 3(b). The DOP is maximum when the film is oriented parallel with the linear polarized beam and reaches minimum for perpendicular orientation therein. The axial angle, \( \phi \), is varied from \(-60°\) to \(60°\) in Fig. 4.
reaction in the DOP becomes significant with decreasing tilting angle. In particular, an increase of more than seven times in the DOP reaction was observed for a tilting angle of 8° compared with the blank substrate. All DOPs were displayed on the sphere extending from the center along the normalized Stokes vector in Fig. 3(b). From Fig. 3 we observed that the responses in DOP and the SOP were indeed significantly affected by the interaction of SWNT with different axial angles. SWNT shows semiconductor characteristics and, when vertically aligned as in Fig. 1(a), SWNT film will produce anisotropy in the interaction with an incident plane electromagnetic wave of a linear polarization.

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

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

In summary, by tilting a vertically aligned SWNT thin film we observed a polarization change at the optical communication window wavelength, 1550 nm, in terms of both the SOP and the DOP in a reproducible manner. A more prominent polarization change was obtained when the SWNT made a shallower angle with respect to the incident direction. In the SOP linear polarization was converted to elliptic polarization, and a response in DOP increased by more than seven times at an axial angle of 8°. Output intensity also depended on azimuthal angle in \( \cos^2 \theta \), consistent with a typical emission of the SWNT.

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