Terahertz waves polarization rotation in photoexcited single-wall carbon nanotube thin film

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Abstract. Terahertz time-domain spectroscopic polarimetry (THz-TDSP) method was used to experimental study polarization properties of unaligned single-wall carbon nanotube thin films with different geometric parameters on transparent float glass substrates in a frequency range from 0.2 THz to 8 THz (corresponding to a wavelength range from ~1.50 mm to ~0.37 mm) at a controlled room temperature of 291–293 K, and a relative humidity of 40–45 %. Frequency dependences of azimuth and ellipticity angles of a polarization ellipse (PE) of electromagnetic waves transmitted through the samples, and PEs at the frequencies of 0.2 THz, 0.5 THz, and 0.8 THz were obtained for values of 0.2 Wcm⁻², 0.6 Wcm⁻², and 1.0 Wcm⁻² of an external 980 nm near infrared optical pumping, with an external static magnetic field of ~0.3 T. Polarization properties were calculated from temporal waveforms of signals transmitted through the samples at the parallel and the crossed by 45° positions to a transmission direction of the polarizers. A change of 15° in the azimuth angle, and of 10° in the ellipticity angle was achieved. The results show that by using carbon nanomaterials-based structures it is possible to devise efficient and affordable magneto-optically tunable polarization modulators that can be used in the advanced areas of terahertz nanoscience and nanotechnologies.

Keywords: terahertz time-domain spectroscopic polarimetry, unaligned single-wall carbon nanotubes, polarization rotation, polarization properties, Stokes parameters, Faraday effect.

1. Introduction

In recent years, there has been a strong improvement in technologies that are aimed at developing methods for generating, detecting and modulating electromagnetic (EM) waves in a terahertz (THz) frequency range. THz EM radiation is widely used to solve fundamental and applied problems of physics, astronomy, chemistry, and medicine [1]. But for the further development of THz technologies, efficient and tunable devices for modulating the polarization, the frequency and the amplitude of EM waves are essential.

In this regard, one of the most important tasks of modern THz photonics is a study of advanced materials, on a basis of which it is possible to create devices with characteristics that exceed all existing analogues. A promising solution to the foregoing problem is a study of carbon nanomaterials-based structures (CNBSs) [2] for use as a functional medium in THz modulators.
CNBSs, including graphene and carbon nanotubes (CNTs), have unique optical and electronic properties, which allows them to be used in the advanced areas of quantum physics, nanophotonics, and nanomedicine.

From a point of view of physics, one of the methods to control the polarization properties of the CNBSs is to use the magneto-optic Faraday effect (MOFE), and the magneto-optic Kerr effect (MOKE). Therefore, recent research is concentrated in a field of studying an influence of an external optical pumping (OP), and an external static magnetic field (MF) on the properties of CNTs, and development of the tunable THz polarizers based on them [3]. For experimental studies of polarization properties of materials, the MOFE, and the MOKE, THz-time-domain spectroscopic polarimetry (TDSP), and THz-time-domain spectroscopic ellipsometry (TDSE) methods are widely used [4]. These methods are an upgrade of the well-known THz time-domain spectroscopy technique and allow obtaining necessary characteristics of studied samples.

But despite the great progress in theoretical and experimental research of CNTs, the influence of the geometric parameters of nanotubes on their optical properties in the THz frequency range has not yet been sufficiently studied, and many knowledge gaps remain in this area.

The goal of this work was an experimental study of unaligned single-wall carbon nanotubes (U-SWCNTs) with different geometric parameters using the THz-TDSP method with an external near infrared (NIR) OP system to obtain their polarization properties.

In this work, polarization properties of the U-SWCNT thin films on transparent float glass (TFG) substrates under the external NIR OP, and the external static MF of ~0.3 T were obtained experimentally in a frequency range from 0.2 THz to 0.8 THz (corresponding to a wavelength range from ~1.50 mm to ~0.37 mm). It was found that with the simultaneous use of the NIR OP of 1.0 Wcm$^{-2}$, and the external static MF of ~0.3 T an azimuth angle of a polarization ellipse (PE) of the EM waves transmitted through the samples changes up to 15°, and the ellipticity angle changes up to 10° relative to the U-SWCNT thin films without influences. This is enough to devise compact magneto-optically tunable polarizers based on U-SWCNTs, which are necessary in advanced areas of THz photonics.

2. Materials and methods
For the study, two U-SWCNTs samples with different geometric parameters were selected. The U-SWCNTs were synthesized by the chemical vapor deposition (CVD) method at KTH Royal Institute of Technology’s Microsystems Technology Laboratory (Kingdom of Sweden) on a nitrocellulose micropore filter (NCMF) using ethanol gas without adding the hydrogen [5]. Then the CNTs were transferred from the NCMF onto the TFG substrates. This substrate was chosen based on the requirement of maximum transmission of THz radiation in the operating range of an experimental setup. The diameters of CNTs were calculated using the Kataura plot [6], the length of the CNTs was visualized by the transmission electron microscopy [7], and the CNTs film thickness was calculated from an optical absorbance at 550 nm [8]. Parameters of the experimental samples are shown in Table 1.
To study a surface morphology of the experimental samples, scanning electron microscopy (SEM) images [9] were obtained at various scales. Based on the SEM images, it can be seen that the CNTs are distributed over the substrates in a form of a disordered mesh with a various brightness, which also means a various density of CNTs. By studying the Raman spectrum, it was found that the samples have a high degree of purity and a small number of structural defects [10].

To study the polarization properties of the experimental samples using the THz-TDSP method [11], a system based on a THz time-domain spectrometer [12], three wire grid polarizers, a 980 nm laser for creating the external OP of 0.2 Wcm$^{-2}$, 0.6 Wcm$^{-2}$, and 1.0 Wcm$^{-2}$, and an axially magnetized NdFeB magnet for creating an external static MF of $\sim$0.3 T were used.

Temporal waveforms of the THz signals transmitted through the experimental samples under the various external influences were recorded using PC with LABVIEW® (National Instruments, Corp., United States of America) software at the parallel and the crossed by 45° positions to the transmission direction of the polarizers [13]. By recording temporal waveforms of transmitted THz signals at two different positions of the polarizers, it is possible to calculate the Stokes parameters and fully describe the polarization properties of the experimental samples.

Each single measurement took $\sim$43 min. All measurements were done at ITMO University’s Terahertz Biomedicine Laboratory (Russian Federation) under a controlled room temperature of 291–293 K, and a relative humidity of 40–45%.

Raw experimental data processing was done using MATLAB® (The MathWorks, Inc., United States of America) software and the Gaussian window function [14]. Experimental samples and setup are shown in Figure 1.

### 3. Results and discussion

Experimental frequency dependences of an azimuth angle $\psi$ and an ellipticity angle $\chi$ of a PE of the EM waves transmitted through the samples in the range from 0.2 THz to 0.8 THz were calculated according to the Equation 1, and Equation 2 [15]:

\[
\begin{bmatrix}
S_0 \\
S_1 \\
S_2 \\
S_3
\end{bmatrix} = \begin{bmatrix}
|E_1|^2 + |E_2|^2 \\
|E_1|^2 - |E_2|^2 \\
2|E_1||E_2|\cos\delta \\
2|E_1||E_2|\sin\delta
\end{bmatrix},
\]

(1)

\[
\begin{bmatrix}
\psi \\
\chi
\end{bmatrix} = \begin{bmatrix}
\frac{1}{2}\arctan\left(S_2\cdot S_1^{-1}\right) \\
\frac{1}{2}\arcsin\left(S_3\cdot S_0^{-1}\right)
\end{bmatrix},
\]

(2)

where $S_0$, $S_1$, $S_2$, and $S_3$ are Stokes parameters; $E_1$, and $E_2$ are complex amplitudes of parallel and perpendicular components of an electric field vector $E$, and $\delta$ is a phase difference between them. Results are shown in Figure 2 (a–f).

To better understand the dynamics of changes in the polarization properties of the experimental samples under the external influences, PEs of the EM waves transmitted through the samples at the frequencies of 0.2 THz, 0.5 THz, and 0.8 THz were calculated [16]. Results are shown in Figure 2 (g–o).

From the obtained experimental results, it is seen that, under the NIR OP of 1.0 Wcm$^{-2}$, and the external static MF of $\sim$0.3 T the azimuth angle changes up to 15°, and the ellipticity angle changes up to 10° relative to the U-SWCNT thin films without influences. The changes in the angles are a result of the MOFE. Also, the complex morphology of the surface of the U-SWCNTs, consisting of the disordered mesh of the CNTs, which was confirmed by the SEM images, has an effect on the modulation of the polarization properties.
Figure 1. SEM images of the U-SWCNTs with scale bars of (a) 10 µm, and (d) 1 µm; (b) scheme of the experimental setup (ADC—analog-to-digital converter; BP—balanced photodetector; DAC—digital-to-analog converter; GTP—Glan–Taylor prism; HW—half-wave plate; IF—PTFE ((C_2F_4)_n) infrared cut-off filter; L1, L2—lenses; LD1, LD2—laser diodes’ drivers; LIA—lock-in amplifier; LR1—1040 nm laser; LR2—980 nm laser; OC—optical chopper; OCC—optical chopper controller; ODL—optical delay line; P1, P3—static polarizers; P2—rotatable polarizer; PC—personal computer; PS—balanced detector power supply; QW—quarter-wave plate; TD—THz radiation detector based on the CdTe crystal; TS—THz radiation source based on the InAs crystal; WP—Wollaston prism); (e) schematic representation of the samples under the external influences; and (c), and (f) images of the assembled experimental setup.
Figure 2. Frequency dependences of (a), (c), and (e) the azimuth angle $\psi$, and (b), (d), and (f) the ellipticity angle $\chi$ of the PE of the EM waves transmitted through the samples; and PEs of the EM waves transmitted through the samples at the frequency of (g), (j), and (m) 0.2 THz, at the frequency of (h), (k), and (n) 0.5 THz, and at the frequency of (i), (l), and (o) 0.8 THz.

Samples without influences:
(a), (b), (g) @ 0.2 THz, (h) @ 0.5 THz, and (i) @ 0.8 THz

TFG substrate
- Sample № 1
- Sample № 2

Sample № 1:
(c), (d), (j) @ 0.2 THz, (k) @ 0.5 THz, and (l) @ 0.8 THz;
Sample № 2:
(e), (f), (m) @ 0.2 THz, (n) @ 0.5 THz, and (o) @ 0.8 THz

- MF of ~0.3 T
- MF, OP of 1.0 W cm$^{-2}$
- MF, OP of 0.2 W cm$^{-2}$
- MF, OP of 0.6 W cm$^{-2}$
4. Conclusion
As a result of this work, the polarization properties of the U-SWCNTs with different geometric parameters in the frequency range from 0.2 THz to 0.8 THz were studied at the controlled room temperature of 291–293 K, and the relative humidity of 40–45%. The change of 15° in the azimuth angle, and of 10° in the ellipticity angle was achieved. It can be seen that the U-SWCNTs is an efficient nanomaterial for device magneto-optically tunable polarizers, which are necessary in cutting-edge THz security and telecommunication systems.

Acknowledgments
The reported study was funded by the Government of the Russian Federation, Grant Number 08–08 (‘Project 5–100’), and by the Russian Science Foundation (RSF), Grant Number 19–72–10141 (‘Thin-film structures based on bismuth and antimony for terahertz photonics’). On behalf of all authors, the corresponding author states that there is no conflict of interest.

Author contributions
A. K.: Conceptualization, Project administration, Data curation, Methodology, Investigation, Formal analysis, Validation, Visualization, Writing—original draft. P. D.: Methodology, Investigation. M. N.: Software. I. A.: Resources. K. B.: Methodology, Investigation. A. B.: Resources, Formal analysis. M. K.: Conceptualization, Supervision, Funding acquisition, Resources, Methodology, Formal analysis, Validation, Writing—review and editing.

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