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研究課題名(和文) Novel terahertz waveguide structure: probing low dimensional materials under extremely strong electromagnetic field

研究課題名(英文) Novel terahertz waveguide structure: probing low dimensional materials under extremely strong electromagnetic field

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研究成果の概要(和文)：標準的な平行平板導波路(PPWG)を用いて、準一次元材料であるカーボンナノチューブ(CNT)の線形テラヘルツ(THz)応答を測定し、従来のテラヘルツ時間領域分光法(TDS)で得られた結果と比較した。従来のTHz-TDSの低THz周波数領域(0.2-0.5THz)吸収は20%であったが、標準PPWGでの吸収は75%に達した。THz-PPWG分光では、特殊形状の試料と特別構成の電場による大きな吸収を利用して、導電率の実部と虚部を高い精度で測定可能となる。また、空気プラズマによる超広帯域THz-TDSシステムを構築した。約10テラヘルツの帯域幅と約1000のS/Nを示している。

研究成果の学術的意義や社会的意義

Investigation of electromagnetic properties of thin film has become of utmost importance with the growing interest in 1D / 2D materials which constitute the building block of future electronic components. We have established an approach to understand the properties properties of these materials.

研究成果の概要(英文)：We have measured the linear terahertz (THz) response of the of a quasi - one dimensional material, namely carbone nanotubes (CNT), with a standard parallel plate waveguide (PPWG) and compared the results with those obtained by the conventional terahertz time domain spectroscopy (TDS). The absorption in the low THz frequency range (0.2-0.5 THz) attained 75% with standard PPWG in the transverse electric mode. In the conventional THz-TDS, the THz absorption is only 20%. In THz PPWG spectroscopy, the huge absorption is due to the special configuration of the sample and the electric field which enables us to measure with a certain accuracy the real and the imaginary part of the conductivity. We have also build an ultrabroadband THz-TDS system based on air plasma. The preliminary result indicates a THz bandwidth about 10 THz with a S/N around 1000.

研究分野：Terahertz photonics

キーワード：terahertz ultrathin materials waveguide spectroscopy strong field broadband

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## 様式 C - 19、F - 19 - 1、Z - 19 (共通)

### 1. 研究開始当初の背景

Low dimensional materials including two dimensional and one dimensional materials such as graphene, monolayer transition metal dichalcogenides (TMD) and carbon nanotubes host new optical and electronic phenomenon. Due to the restriction of the carrier motions to a plane or in line and the reduced bulk dielectric screening, the physics of low dimensional system are fundamentally different from their bulk counterpart. Although the optical (visible and infrared) and electronic properties of these novel atomic layer structures are well investigated, two most fundamental questions still remain for a real world application. On one hand, charged carrier (electrons, holes, charged excitons, plasmons) dynamics in the low dimensional material are yet to be fully explored and explained. The transport mechanism of these carriers dictate the performance of low dimensional-based optoelectronic devices (Nanophotonics **6**, 1309 (2017)). Few investigations have recently started to elucidate the carrier transport mechanism in sub-picosecond scale of 2D/1D materials using the conventional scheme in terahertz (THz) time domain spectroscopy (TDS) (ACS Nano **8**, 1147 (2014)). However, the lack of the low conductive atomic layer response to THz radiation renders this standard method less ideal, especially for the study of semiconducting low dimensional materials. Therefore, the main challenge is to establish an effective, reconfigurable, nondestructive spectroscopic modality which is compatible with the state of the art of the research.

### 2. 研究の目的

This project entitled “Novel terahertz (THz) waveguide structure: probing low dimensional materials under extremely strong electromagnetic field” aims to probe linear and nonlinear THz responses of low dimensional materials including graphene, carbon nanotubes (CNT), molybdenum disulfide ( $\text{MoS}_2$ ). The novelty proposed in the project is the use of a new parallel plate waveguide (PPWG) structure as shown Figure 1. In fact, the waveguide concept is based on the cross propagation of the THz pump and THz probe. This PPWG geometry permits to pump the low dimensional samples under test with an extremely high power THz radiation and subsequently to probe their nonlinear THz responses.

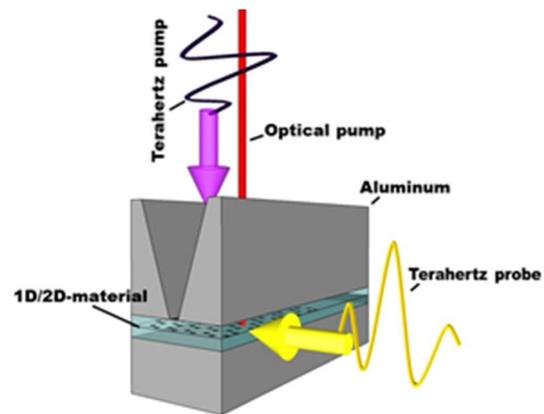


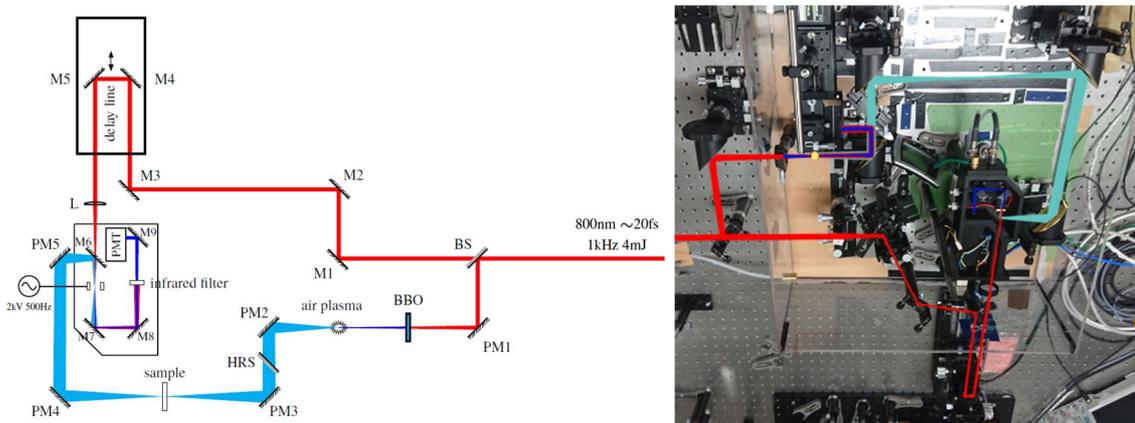
Figure 1 Waveguide Structure

### 3. 研究の方法

The first step of the project was to probe the linear THz responses of the 1D/2D materials. We have used conventional THz - TDS transmission type system and standard PPWG THz - TDS system. The conventional transmission THz-TDS is composed of a THz emitter, THz detector and a system of 4 parabolic mirrors to shape the THz radiation (Nat. Photonics **1**,97-105(2007)). For the PPWG THz - TDS system, we use a bare p-type indium arsenide wafer emitter excited by a Ti: sapphire laser at 800 nm and 80 MHz repetition rate. A photoconductive dipole antenna on gallium arsenide detects the THz wave. The waveguide is composed of two flat aluminum plates with 1 mm plate separation, 10 mm length, and 30 mm width. The PPWG is placed in the confocal beam waist of four off-axis parabolic mirror system in order to have a nearly frequency-independent focused wave at the waveguide input facet (J Infrared Milli Terahz Wave **36**, 1182-1194 (2015)).

The second step of the research was to build a broadband THz - TDS system based on air plasma. As shown Figure 2, The broadband THz spectrometer is driven by the Ti: sapphire amplified laser system (Spitfire Pro by Spectra-Physics) producing ultrashort laser pulses of 100 fs duration at 800nm and at 1 kHz repetition rate. The 4 mJ laser pulse energy is split into two parts respectively: 3.6 mJ for the THz generation, and 0.4 mJ for the THz sampling. Here, the THz wave is generated by two color mixing air plasma. A parabolic mirror with 152.4 mm focal length focus the pump laser beam in the air.

Then, a 100  $\mu\text{m}$ -thick  $\text{-BBO}$  crystal, for second harmonics generation, is mounted on a linear stage at 5 cm before the focal point of the parabolic mirror. The THz radiation emitted by the plasma filament is shaped and focused by a 4f system of parabolic mirrors to form the THz - TDS. Before entering an ABCD detector, the average pulse energy of the probe beam is attenuated to  $\sim 60 \mu\text{J}$ , and is focused between the high voltage electrodes located inside of the ABCD detector by a 150 mm-focal length lens.

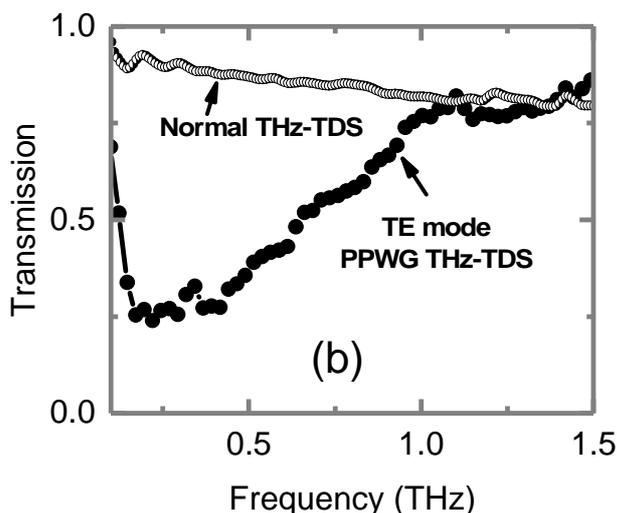


**Figure 2** Schematic a) and actual picture b) of the ultra-broadband THz setup based on THz air photonics

When an AC bias voltage is synchronized with the lock-in amplifier, we can acquire the entire THz time waveform by sampling the THz pulse as a function of the delay collected by scanning the probe delay line between the THz and probe pulses.

#### 4 . 研究成果

We have performed THz waveguide spectroscopy experiments with few samples including arc discharged carbon nanotubes (1D material) and chemically vapor deposited (CVD) graphene. The arc discharged carbon nanotubes samples have no defined electrical conductivity but rather a mixture of conductive and semiconducting materials. The CNT samples are deposited on high resistive silicon and magnesium oxide ( $\text{MgO}$ ) substrates. The dimensions of the substrates are 30 mm of width, 10 mm of length and 0.5 mm of thickness. One set of CNT samples is aligned parallel to the width of the substrates and another set is aligned parallel to the length of the substrates. A different set of CNT samples is just nonaligned, i.e. the tubes are randomly oriented during the deposition process. These samples, including CVD graphene, are prepared in collaboration with Rice University. By using a simple parallel plate waveguide (PPWG), we are able to determine the THz sheet conductivity of the samples. In the THz PPWG spectroscopy configuration, the thin film samples are located in-between of two block of aluminum. The electric field of an incoming THz wave is polarized either parallel or perpendicular to the surface of the samples. When the electric field is parallel to the samples surface, we are in transverse electric (T.E.) mode of the PPWG.

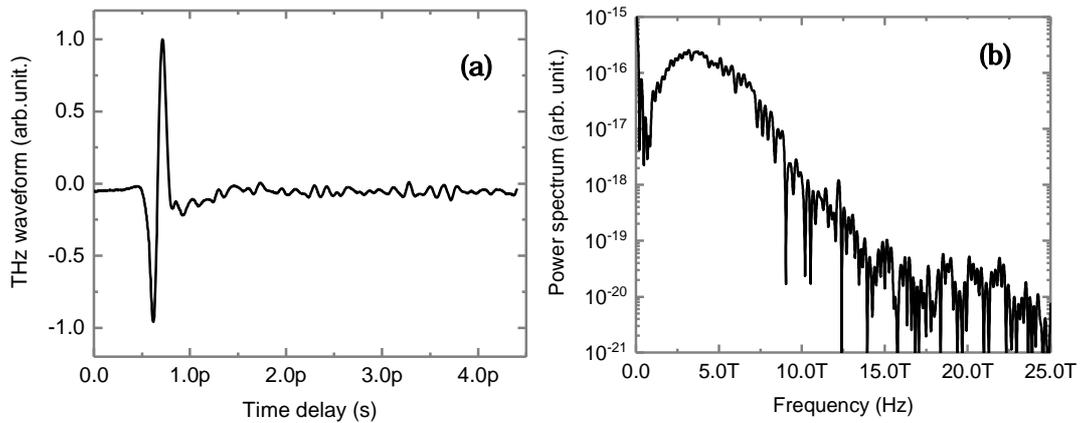


**Figure 3** Comparison of transmission spectrum between conventional THz - TDS and PPWG - THz - TDS in the TE mode for unaligned CNT on Si substrate.

When the electric field is perpendicular to the samples surface, we are in transverse magnetic (T.M) mode of the PPWG. In the case of CNT samples, the conductivity is not isotropic and thus depends on direction of the electric field. When the electric field

is normal to the sample surface, *i.e.* in T.M mode, the response of the materials is extremely weak since the motion of the carriers are confined in sub-nanometer scale in that direction. Therefore, the detection of this material response in TM mode is beyond the limit of the system. In T. E mode, the electric field of the incoming THz wave is parallel to the surface and the direction of the CNT. Here, the THz absorption can reach a huge amount for an extremely thin samples. For example, as shown Figure 3, for unaligned CNT on Si substrate, the absorption in the low frequency region (0.2 - 0.5 THz) is 75% in THz waveguide spectroscopy compared to 20% for the standard method of THz spectroscopy. The use of the PPWG spectroscopy has been revealed to be advantageous in our previous research with single atomic layer and has been confirmed in this current research. In the standard THz - time domain spectroscopy (TDS), this weak light - matter (1D/2D materials) interaction limits the determination of the conductivity. In such case, only the real part of the conductivity is reliable and is usually fitted with the simple Drude conductivity model for the quasi-metallic CNT as well as for the graphene samples. In THz PPWG spectroscopy, the huge absorption due to the special configuration of the sample and the electric field enables to measure with a certain accuracy the real and the imaginary part of the conductivity. The striking difference with the standard THz - TDS is that the Drude conductivity roll-off in the waveguide measurement occurs at lower frequency. This implies that the carrier relaxation time is one to two magnitude slower than the previous results published in the literatures for the case of CNT or single layer graphene.

For the broadband THz - TDS system based on THz air photonics, we have obtained a THz spectrum with bandwidth of 6 THz and improved it to 12 THz bandwidth. The signal to noise ratio (S/N) of the THz spectrum was about 200 in the first step. Here, the goal is to reach a spectrum of about 12 THz bandwidth with a S/N around 1000 since the waveguide generally transmits 20% of the input signal. Currently, we have improved the laser system to generate a pulse duration of 35 fs by optical fiber compression of the original 100 fs laser pulse. The preliminary results indicate a THz spectrum of 10 THz with a S/N around 1000 as shown Figure 4.



**Figure 4** (a) Time domain waveform and (b) Frequency domain of the THz air photonics pulse.

5 . 主な発表論文等

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3 . 学会等名 The 67th JSAP Spring Meeting 2020
4 . 発表年 2020年

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〔図書〕 計0件

〔産業財産権〕

〔その他〕

Ono/Hori lab web site  
<https://wpp.shizuoka.ac.jp/nano/>

6. 研究組織

	氏名 (ローマ字氏名) (研究者番号)	所属研究機関・部局・職 (機関番号)	備考
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