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科研費
KAKENHI

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研究課題名（和文）Development of three-dimensional terahertz photonic chip for molecular sensing

研究課題名（英文）Development of three-dimensional terahertz photonic chip for molecular sensing

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研究成果の概要（和文）：金属ロッドアレイ(metal rod array: MRA)および多孔質ポリマー構造(microporous polymer structure: MPS)の二種類のフォトニック構造により、三次元フォトニック結晶技術は首尾よく開発が進められている。この技術の応用として、異なる双極子モーメントを持つ液体とガスの分子センシングが実証された。MPSは揮発性有機化合物に対して敏感であり、アセトンの分子量に関しては1ppmまで検出可能である。さらに本研究では、テラヘルツ波を三次元構造に対して相互作用させる基礎研究に基づき、周波数依存性を持つ侵入深さを0.1~1 THzの範囲で示した。

研究成果の学術的意義や社会的意義
The projects realize 3D printing methods in terahertz optics to guide the college students in the University of Tsukuba. The optical sensing methods are also attractive and make minute amounts of molecules visible via THz wave detection. The sensors are useful for the scientists and engineers.

研究成果の概要（英文）：Three-dimensional photonic chips are successfully developed based on two kinds of photonic structures, which are the metal-rod-array(MRA) and microporous-polymer-structure (MPS). The molecular sensing for liquids and gases with different dipole moments are successfully demonstrated on the 3D photonic chips. The MPS photonic chip is sensitive to the vapors of volatile-organic-compounds (VOCs) and the detectable molecular amount is down to 1 ppm for the acetone molecules. The MRA chip is sensitive to the liquids of VOCs, and the detectable molecular amount is down to 0.1 mmole for the dipole-moment difference around 0.01 Debye. Such high sensitivities of the molecular sensing based on THz photonic chips are resulted from the enhanced optical-path-length inside the chips. Based on the theory of terahertz waves interaction with the 3D photonic structures, the research additionally approves the multiple layer model of skin and obtains the frequency-dependent penetration depth in 0.1~1 THz.

研究分野：光工学と光量子科学

キーワード：terahertz waves　photonic structure　optical sensor　terahertz sensing　slab waveguide　terahertz optics　integrated optics　terahertz spectroscopy

１．研究開始当初の背景

To develop a photonic chip, sensing molecules by the terahertz (THz) photons in the near field of three-dimensional (3D) structures, is the research purpose. However, it is not easy to handle in general. The laboratory operations in small scales are thus urgently requested, which are usually known as the lab on a chip and a photonic chip, individually, for the sample and photon manipulations. Using the periodic structures of the photonic crystals and metamaterials as artificial waveguide media is popular, and the 3D constructions are the state of art to explore. Instead of the nano-scaled 3D constructions in the visible/infrared (IR) rays, the size of a 3D structural unit in a THz band is about tens or hundreds of micrometers that are easy to prepare by 3D printing. The THz photonic sensors in the research are comparatively cheap and disposable in the label-free sensing work of analytical chemistry.

２．研究の目的

The research applies terahertz (THz) photon to sense the molecules because of the energy matching to the intermolecular attraction energy. Based on the designed three-dimensional (3D) microstructures, the interaction between THz photons and analyte molecules can be realized on a miniaturized chip with high sensitivity. The research purpose is to explore one 3D-microstructure, possessing multiple functions of a low-loss waveguide, a broadband interferometer and a high quality-factor resonator. The project achievement eventually can study biological samples with multiple layered structures.

３．研究の方法

To achieve molecular sensing, the principal investigator (PI) goes through two main schemes, including a microporous polymer structure (MPS) and a metal rod array (MRA).

(1)　3D THz photonic chips based on a microporous polymer structure (MPS):

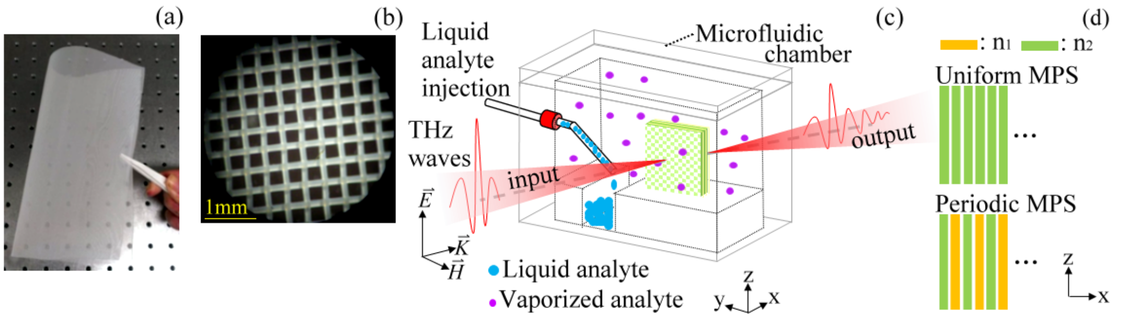

Fig. 1. (a) Photograph of a polyethylene terephthalate (PET) mesh. (b) Microscopic photograph of a PET mesh. White lines: PET grids. Black square holes: Micropores. (c) Configuration of a MPS volatile gas sensor. (d) Uniform and periodic configurations of the MPSs as sketched in the x-z plane.

A MPS volatile gas sensor is constructed by multiple layers of polyethylene terephthalate (PET) mesh (SEFAR PET1000, SEFAR AG, Switzerland) and one microfluidic chamber made of Teflon material. A flexible PET mesh [Fig. 1(a)] consists of periodical square holes, which are two-dimensionally arranged in square arrays as shown in Fig. 1(b). Figure 1(c) shows the schematic diagram of the MPS-based THz volatile gas sensor, which consists of a MPS device and a Teflon sample chamber. To study the structure-dependent sensing performance of MPS, four kinds of PET meshes with different thicknesses and square micropore sizes are utilized to stack into two configurations of MPSs, i.e., uniform and periodic structures [Fig. 1(d)].

(2)　3D THz photonic chips based on a metal rod array (MRA):

Figure 2(a) shows the configuration of a MRA and its microscopic photograph at the inset. The structural period (Λ) is approximately 0.62 mm along the x- and z-axes. The diameter and interspace of metal rods are approximately 0.16 and 0.46 mm, respectively. The length of each rod is almost consistent (approximately 1 mm), and the structural total width along the x-axis is 12Λ. Figure 2(b) illustrates the input and transmitted THz waveforms through the MRAs with different layer thicknesses measured by the waveguide-based THz-TDS. When THz waves pass though the four-layered MRA, the electric field amplitude is severely decayed by the metal rods so that it cannot provide sufficient signal change for liquid fluid sensing.

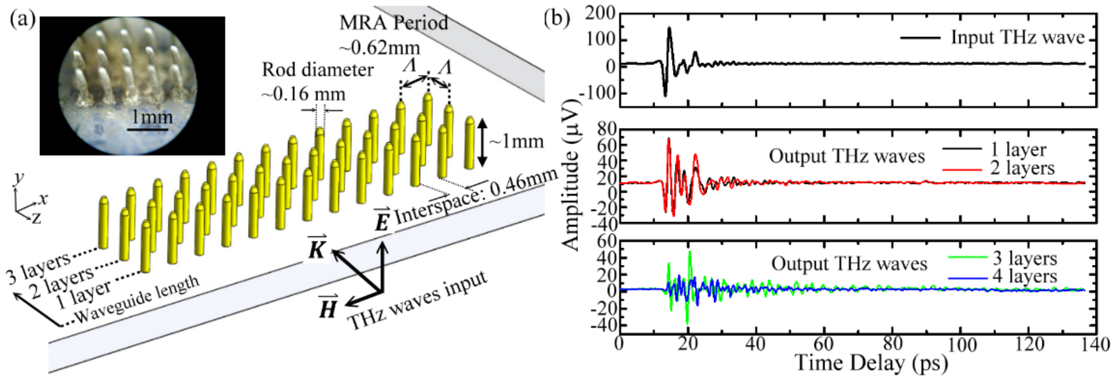

Fig. 2 (a) Schematic configuration of MRA to pass THz waves. (Inset) Microscopic photograph of a MRA. (b) Input and output THz waveforms for the MRAs with different transmission thicknesses.

４．研究成果

The research achievements involve three sections:(1)3D THz photonic chips based on a microporous polymer structure, (2) 3D THz photonic chips based on a metal rod array, and (3) biomedical application. For each section, the principal investigator (PI) has finished related topics to approach the development of three-dimensional terahertz photonic chip for molecular sensing.

(1) 3D THz photonic chips based on a microporous polymer structure (MPS):

Topic ①. Sensitivity analysis of multilayer microporous polymer structures for terahertz volatile gas sensing:(The research topic is presented in *Optics Express 25, 5651-5661 (2017).*)

We successfully demonstrate the feasibility of in situ and label-free detection for the VOC and high THz-absorbed volatile liquids using a simple multilayer-stacked MPS. The dependency of vapor sensing ability on the geometrical parameters of the MPS-based THz gas sensor is experimentally verified based on the analysis of THz absorption and refractive index variation before and after acetone vapor infiltration in the MPS structure. We have experimentally proven that the MPS with the smallest micropore volume can efficiently enhance THz absorptions both in the vapor-filled pore-space and on the vapor-adsorbed hydrophilic-pore-surface, resulting in the best sensitivity of approximately 1 ppm to detect a wide concentration range (<200 ppm) of acetone vapor. To our knowledge, the demonstrated sensitivity for VOC detection using the MPS is the highest compared among the resonator-type sensors in the THz regime. Different concentrations of toxic methanol adulterated in alcoholic aqueous solutions are successfully identified using the MPS with the best sensitivity. The MPS device is advantageous for THz wave gas sensing because of its room temperature operation, easy availability, low cost, low THz insertion loss, and label-free VOC sensing capability. The microporous THz gas sensor is highly promising for the detection of environmental pollutants, hazardous gas, and toxic adulterated beverages.

(2) 3D THz photonic chips based on a metal rod array (MRA):

Topic ①. Terahertz artificial material based on integrated metal-rod-array for phase sensitive fluid detection: (The research topic is presented in *Optics Express 25, 8571-8583 (2017)* and specially selected in *Virtual Journal for Biomedical Optics (VJBO), Vol. 12, Iss. 6 (2017).*)

We have experimentally demonstrated a MRA-based THz artificial material to transport the transverse-electric (TE) waves resulted from the resonance among rod slits. The miniaturized MRA-based waveguide, fabricated by the high accuracy of the 3D printing technique, is also successfully incorporated with the microfluidics to transport THz waves and fluidic analytes in the same channel for THz optofluidic sensing applications. Analysis of the THz transmission spectra for different layers of MRAs shows the optical-path-length (OPL) of the MRA-guiding wave is longer than the physical waveguide length. The three-layered MRA is thus suggested as the optimal waveguide sensing length because of the sufficiently long OPL, adequate transmission power and high overlapping field to enhance the wave-analyte interaction. The acetone vapor sensing result shows that the phase variation, rather than the amplitude decay, of the THz field can be strongly enhanced using the 2nd resonance waveguide mode, compared with the sensing result of blank parallel-plate waveguide (PPWG). Finally, the composite MRA-based sensing unity is utilized for liquid sensing based on the phase-sensitive property of the TE guiding waves. With the optimal overlapping between the modal field and liquid overlayer along the rod axis, three types of colorless liquid analytes are successfully recognized at microliter volumes based on their distinct molecular dipole moments. This work demonstrates the realization of a miniaturized THz waveguide using a MRA-based artificial material, and the feasibility of label-free microfluidic detection based on the waveguide sensing platform to distinguish minute amounts of liquid analytes with similar dipole moments. Using the phase-sensitive method for minute liquid detection is not restricted to the broadband THz radiation and the uniform sample cell thickness, which are usually adopted for sensing the absorption lines of materials. The MRA waveguide can be potentially integrated into the micro-total-analysis system as one unit or conjugated with lab chip technology for various sensing applications.

Topic ②. Investigation of spectral properties and lateral confinement of THz waves on a metal-rod-array-based photonic crystal waveguide: (The research topic is presented in *Opt. Express 26, 15570-15584 (2018)*.)

Laterally confined THz wave guidance is experimentally demonstrated on a 1 mm-thick planar waveguide composed of MRA, and the spectral properties of the MRA-based waveguide are comprehensively investigated by using THz transmission spectroscopy and compared with theory. A MRA structure is characterized as one THz-photonic crystal (PC) waveguide, spatially modulating and strongly reflecting THz waves to form one photonic bandgap at 0.10-0.60 THz. The photonic bandgap of an MRA-PC waveguide can be manipulated by changing the MRA geometry parameters and is effectively consistent between experiment and finite-difference time-domain (FDTD) simulation. Increasing the rod diameter and layer number of a MRA structure or decreasing the interspace between adjacent rods obviously increases the spectral width and power distinction (i.e., visibility) of the stop band because the reflectivity of each MRA layer rises to facilitate constructive interference of THz reflection. The power ratio enhancement relating to a spectral peak value for the measured transmission band of 30-layered MRA PC waveguide is verified because of longitudinally resonant guidance of transverse magnetic (TM)-polarized waveguide modes along the MRA length, which is critical to the interspace width of adjacent rods and the metal coating of rod surface. A MRA slab waveguide is further validated to guide both single- and high-order modes along the rod axis (or Z-axis) with different propagation losses. Single waveguide modes of a MRA structure can propagate for long distances with low scattering loss and indicate lateral confinement with power peak at MRA-tip surface because the fractions of modal power inside the MRA slab waveguide are relatively lower than those of the high-order modes. For example, the highest transmission power of a 30-layered MRA waveguide with a 0.26 mm gap-size and a 0.16 mm diameter (D) occurs at around 0.505-0.512 THz. Such MRA-guided THz waves in this frequency range (0.505-0.512 THz) are experimentally and theoretically demonstrated as single-mode guidance with modal power peaks laterally confined on the metal rod tips and the corresponding fractional power inside MRA is approximately 40%. Based on the ideal assumption (i.e., perfect electric conductor), 0.505-0.512 THz waves indicate extremely low scattering loss, 0.003 cm^{-1}, and propagate for a long distance over 30 mm with the transmittance above 30%.

Topic ③. Geometry-dependent modal field properties of metal-rod-array-based terahertz waveguides: (The research topic is presented in *OSA Continuum 2, 655-666 (2019)*.)

The MRA structure is numerically demonstrated by FDTD calculation as one THz waveguide. The fundamental and high-order TM waveguide modes are distinctly found in 0.1-1 THz along the 30-layer MRA propagation. For the uniform metal rod interspace, the high-order waveguide modes divided by the Bragg-like reflection originate from the sufficiently long metal rod length. For 1-mm-long rod in an MRA, assembling the metal rods with an asymmetric interspace (i.e., $G_x \neq G_y$) is presented as the critical stratagem to tailor the high-order TM waveguide mode with optimal transportation efficiency of THz waves. The G_x and G_y respectively denote the interspace of metal rod along the X- and Y-axes, perpendicular to the rod axis, i.e., Z-axis. We consider the 0.16 mm D, 0.26 mm G_y, and 1-mm-long rod length as the geometrical basis to adjust G_x in 0.08-0.50 mm and find apparent spectral redshift by increasing G_x for the MRA-TM modes. Such a spectral shift that is inversely proportional to the G_x interspace resembles the PPWG performance on the TM waveguide modes that depend on the hollow core spaces. The G_x interspace channel constructed by two MRA lines is thus the structural unit for determining the modal properties of TM-THz waves based on the effective medium concept. On the basis of 0.16 mm D, 0.26 mm G_y, and 1-mm-long rod, the 2nd TM waveguide mode through 30-layer MRA propagation is stably performed in 0.4-0.7 THz for 0.08-0.50 mm G_x. Based on the observation of G_x-dependent modal field properties, the MRA-guided THz field is exactly bound on the metal rod surface with the lowest waveguide loss or the high transmittance; thus, when G_x approaches 0.20 mm, the efficiency of THz waveguide, which is defined by the transmittance, bandwidth, and attenuation, can be optimized. The longest waveguide length and widest bandwidth can consequently be obtained when the symmetric interspace of 0.26 mm is modified as the asymmetric one with 0.20 mm G_x and 0.26 mm G_y. The natural confinement performance of the 2nd TM mode at the MRA-air interface enables the MRA structure to be feasible for a THz slab dielectric waveguides.

(3) Biomedical application:

Topic ①. **Frequency-dependent skin penetration depth of terahertz radiation determined by water sorption-desorption**:(The research topic is presented in *Optics Express 26, 22709-22721 (2018).*)

The THz frequency-dependent property of skin penetration depth is experimentally identified during water sorption-desorption on a skin sample. For water sorption, the multilayered water-skin medium is constructed to observe THz wave interference effect by placing a water drop on the sample skin surface. Water desorption is considered as the natural evaporation of water under ambient atmosphere until the skin surface is dry. The multilayered water-skin structure is continuously irradiated with THz waves during water evaporation to dynamically record the reflected amplitudes of THz waves. Skin samples under water overlayer thicknesses of more than 0.4 mm and the dry condition can be modeled as a multiple layer configuration with weak interference of THz wave. The THz refractive indices and absorption coefficients of the weak interference condition can thus be derived via the FEM-based iterative approximation of THz reflectivity. Skin under water overlayer thicknesses of less than 0.2 mm but larger than 0 mm can be modeled as a multiple layer configuration with strong interference of THz wave. The THz interactive layer thicknesses correspond to THz penetration depth and are consequently obtained through the iterative approximation of finite-element method (FEM) calculation of THz reflectivity. The sensing results show that the 0.1-0.9 THz waves have skin penetration depths of 0.1-0.3 mm and those waves with 0.4-0.6 THz frequencies especially have the maximum skin penetration depth of 0.3 mm. To confirm the maximum penetration depths of THz waves and to validate the multilayered skin model, the skin surface is further damaged with boiling water and freezing at -85 ° C to induce massive membrane disruption, thereby forming a large porous space in the skin surface with a high water content. The porosity or water content of the damaged skin tissue with a thickness of 0.1 mm is nearly 80% and thus approximates a water-like tissue layer at the skin surface. When THz waves reflect from the damaged skin, the waves with frequencies of >0.6 THz or <0.38 THz possibly have penetration depths of less than 0.27 mm and could be blocked by the outermost damaged tissue, thereby performing high reflected field fluctuation during the water loss. Contrarily, the reflected field variation of THz waves with frequencies at 0.383, 0.460, and 0.536 THz are linearly related with water loss, which probably results from the penetration depths of more than 0.27 mm. In contrast to that of 0.383 and 0.460 THz waves, the amplitude variation of 0.536 THz wave is inversely proportional to the water loss amounts given the constructive interference between the first and second reflections at the outermost water layer and innermost hydrated skin tissue, respectively.

５．主な発表論文等

〔雑誌論文〕（計６件）

The following papers are all refereed.

1. Dejun Liu, Ja-Yu Lu, Borwen You*, and Toshiaki Hattori, "Geometry-dependent modal field properties of metal-rod-array-based terahertz waveguides," OSA Continuum 2, 655-666 (2019). (https://doi.org/10.1364/OSAC.2.000655) (*: corresponding author)
2. Borwen You, Ching-Yu Chen, Chin-Ping Yu, Pei-Hwa Wang, Ja-Yu Lu, "Frequency-dependent skin penetration depth of terahertz radiation determined by water sorption-desorption," Optics Express 26, 22709-22721 (2018). (https://doi.org/10.1364/OE.26.022709)
3. Borwen You, D. Liu, T. Hattori, T.-A. Liu, and J.-Y. Lu, "Investigation of spectral properties and lateral confinement of THz waves on a metal-rod-array-based photonic

crystal waveguide," Opt. Express 26, 15570-15584 (2018).

4. C.-K. Sun, H.-Y. Chen, T.-F. Tseng, Borwen You, M.-L. Wei, J.-Y. Lu, Y.-L. Chang, W.-L. Tseng and T.-D. Wang, "High Sensitivity of T-Ray for Thrombus Sensing," Sci. Rep.-UK 8, 3948 (2018). (DOI:10.1038/s41598-018-22060-y)
5. Borwen You, Ching-Yu Chen, Chin-Ping Yu, Tze-An Liu, Toshiaki Hattori, and Ja-Yu Lu, "Terahertz artificial material based on integrated metal-rod-array for phase sensitive fluid detection," *Optics Express* 25, 8571-8583 (2017). (https://doi.org/10.1364/OE.25.008571)(*Selected in Virtual Journal for Biomedical Optics (VJBO), Vol. 12, Iss. 6 (2017)*)
6. Borwen You and Ja-Yu Lu, "Sensitivity analysis of multilayer microporous polymer structures for terahertz volatile gas sensing," *Optics Express* 25, 5651-5661 (2017). (https://doi.org/10.1364/OE.25.005651)

〔学会発表〕（計 **7** 件）

The following items only list oral presentations by the principal investigator (PI).

1. Borwen You, Ja-Yu Lu, Chin-Ping Yu, and Toshiaki Hattori, "Miniature terahertz waveguides and the molecular sensing application in a microfluidic channel," The 66th Japan Society of Applied Physics (JSAP) Spring Meeting, paper 11p-S421-16, Tokyo (Tokyo Institute of Technology, Ookayama Campus), Japan 2019 (2019/3/9~12).
2. Borwen You, Ja-Yu Lu, Chin-Ping Yu, Pei-Hwa Wang, "Functional assessment of water sorption‐desorption for frequency-dependent skin penetration depth of terahertz radiation," The 66th Japan Society of Applied Physics (JSAP) Spring Meeting, paper 12a-S421-5, Tokyo (Tokyo Institute of Technology, Ookayama Campus), Japan 2019 (2019/3/9~12).
3. Borwen You and Ja-Yu Lu, "Terahertz artificial material based on integrated metal-rod-array for phase sensitive fluid detection," 43th Infrared, Millimeter and Terahertz Waves (IRMMW-THz), paper Th-A2-1b-4, Nagoya, Japan 2018 (2018/9/9~9/14).
4. Borwen You, Ja-Yu Lu and Toshiaki Hattori, "Terahertz integrated waveguide sensor based on a metal rod array for phase sensitive fluid detection," Progress in Electromagnetics Research Symposium (PIERS), session 3P2b, Nanyang Technological University, Singapore 2017 (2017/11/19~22).
5. Borwen You, Ja-Yu Lu, and Toshiaki Hattori, "Terahertz volatile gas sensing and sensitivity analysis based on microporous polymer structures," 42th Infrared, Millimeter and Terahertz Waves (IRMMW-THz), paper RB 1.1, Cancun, Mexico 2017 (2017/8/27~9/1).
6. Borwen You, Ja-Yu Lu and Toshiaki Hattori, "Optical sensing development based on THz fibers," 2016 Symposium of terahertz science cutting edge III, Fukui (Sakai), Japan 2016 (invited talk).
7. Borwen You, Ja-Yu Lu and Toshiaki Hattori, "Chemical remote and in situ sensing based on a flexible terahertz pipe waveguide," The 5th joint symposia with the Optical Society of America (OSA) in the 77th Japan Society of Applied Physics (JSAP) Autumn Meeting, paper 15p-C301-11, Niigata, Japan 2016.

〔図書〕（計 **2** 件）

1. Borwen You and Ja-Yu Lu, "Integrated terahertz planar waveguides for molecular sensing," Chapter 8 in *Advances in Optics: Reviews (Vol.3)*, Sergey Y. Yurish (IFSA Publishing - International Frequency Sensor Association, 2018), pp. 197-212. (The page amount of the book is 522.)
2. Borwen You and Ja-Yu Lu, "Terahertz Fiber Sensing," Chapter 4 in *Terahertz Spectroscopy-A Cutting Edge Technology*, Jamal Uddin (InTechOpen, 2017) pp. 63-81. (DOI: 10.5772/66345) (The page amount of the book is 307.)

〔その他〕

Other conference publications or any update achievement could be reviewed in the PI's home page, University of Tsukuba - researchers information, or http://www.trios.tsukuba.ac.jp/researcher/0000003718

６．研究組織

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