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研究課題名（和文）原子オーダー熱探査プラットフォームと熱の波動性の解明

研究課題名（英文）Platform for atomic-order thermal probing and elucidation of wave-like heat conduction

研究代表者
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研究成果の概要（和文）：本研究では、ラマン分光と走査型電子顕微鏡に基づいた高分解能を持つ熱物性計測法を開発した。 まず、ラマン分光に基づく温度計測の分解能を向上させた。 また、一次元材料の熱伝導率と内部構造の同定手法を開発し、電子線加熱を駆使して高分解能な熱抵抗マッピング手法も確立した。 さらに、フォノンの波動性を解明するために、グラフェンにおいて超微細なナノ構造を加工することに成功した。

研究成果の学術的意義や社会的意義
本研究にて得られた成果は、ナノスケール熱物性の新たな計測方法を開発することで、熱物性分野の発展に貢献した。本研究で開発した熱計測法はナノスケール伝熱機構の解明への応用を期待でき、電子機器の高度な熱管理技術の開発および高度な情報化社会の達成へ貢献できる。

研究成果の概要（英文）：Throughout this project, we developed high-resolution thermal property measurement methods based on Raman spectroscopy and scanning electron microscopy. We improved the temperature resolution of Raman-based thermal measurements. We also developed a method to simultaneously measure the thermal conductivity and observe the internal structure of 1D materials and introduced electron-beam heating for high-resolution thermal resistance mapping. In addition, we successfully fabricated ultrafine nanostructures in graphene to study phonons’ wave behaviors.

研究分野：熱工学

キーワード：ナノスケール熱物性

１．研究開始当初の背景

In recent years, due to the miniaturization and high-speed performance of electronic devices, thermal issues such as ultra-high heat flux, hotspots, and interfacial thermal resistance in the micro- and nanoscale have been predicted to become a bottleneck for the sustainable development in the super-smart society (Waldrop M. Nature, 2016). With the development of nanomaterials and nanostructures for advanced heat control underway, there still lacks a universal and highly precise thermophysical measurement platform for the nanoscale thermal property evaluation. In addition, although there has been growing attention to the wave nature of phonons (i.e. quasi-particles of heat conduction) in nanostructures, the experimental elucidation of the wave-like behaviors of heat is still very limited.

２．研究の目的

This research project aims to develop a high-resolution thermal property measurement platform, and experimentally elucidate the wave-like behaviors of heat.

３．研究の方法

We utilized Raman spectroscopy and scanning electron microscopy to develop high-resolution thermal measurement techniques and fabricated graphene nanostructures for the study of wave-like heat conduction.

４．研究成果

First, we have established the lock-in Raman technique to greatly improve the temperature resolution of Raman thermometry. One of the biggest challenges of Raman thermometry is the relatively low temperature resolution, which is usually larger than 5 K. Here, we use modulated electrical current at a low frequency to heat the sample and simultaneously collect the Raman shifts that are linearly related to temperature. Then, we treat the Raman shifts data with fast Fourier transform and pick out the Raman shift with the same frequency as the heating current, so that the noises at other frequencies can be readily rooted out. Using this lock-in Raman thermography technique, we have successfully measured the thermal contact resistance between individual carbon fibers with high accuracy. We have also combined the MEMS sensor with the lock-in Raman technique for thermal resistance mapping along an individual nanotube.

Second, we have established an experimental method that allows concurrently the in-situ thermal conductivity measurement and the real-time internal structure observation of a single one-dimensional (1D) material using scanning transmission electron microscopy in a scanning electron microscope (STEM-in-SEM) (Fig. 1). Using this method, we *in-situ* measured the thermal conductivities of individual cup-stacked carbon nanotubes and concurrently observed the internal hollow structures. We found that the sample with more structural disorders had a lower thermal conductivity. Our measurement method can pave the way to the sample-by-sample elucidation of the structure-property relationship for 1D materials. This achievement has been published in Appl. Phys. Lett. 2022, 120(4), 043104.

Third, we combined the electron beam (EB) heating with two suspended line-shaped heat flux sensors and achieved the *in-situ* thermal resistance mapping along a single carbon nanotube (CNT) in a scanning electron microscope with a nanometer-range spatial resolution (Fig. 2). The CNT is anchored between the two suspended metal lines, and the focused electron beam heats the CNT locally with an ultrahigh spatial resolution, while the two metal lines simultaneously measure the heat fluxes induced by the EB heating. We can obtain the spatially resolved thermal resistance by sweeping the focused EB along the CNT. Utilizing this technique, we extracted the thermal conductivity of the CNT without the errors from the thermal contact resistance between the sample and the sensors. This achievement has been published in Int. J. Heat Mass Transf. 2022, 198, 123418, and paved the way for the experimental elucidation of various localized heat conduction phenomena, including the wave effect.

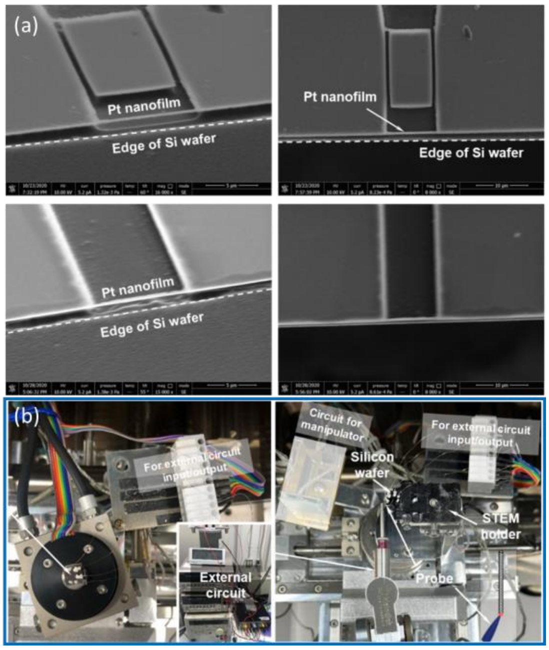

Fig. 1 Experimental system for the concurrent thermal conductivity measurement and internal structure observation of 1D materials. (a) SEM images of the suspended Pt sensor on the wafer edge; (b) photos of the experimental setup for the *in-situ* STEM-in-SEM measurements.

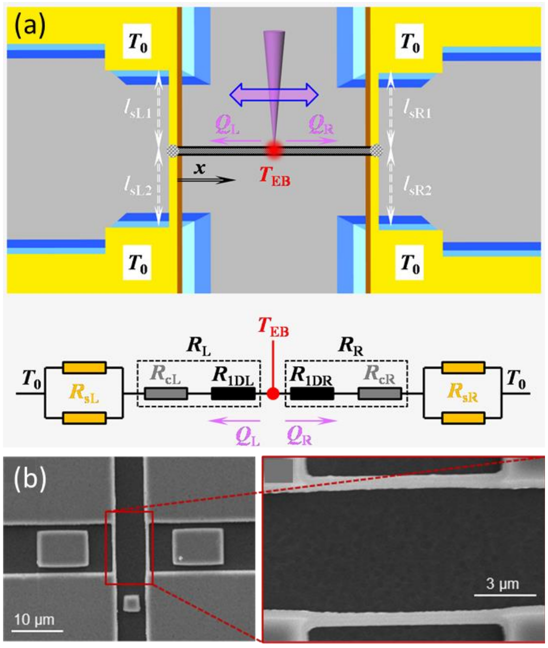

Fig. 2 (a) Schematic diagram and the corresponding equivalent thermal circuit of the thermal resistance mapping measurement using the focused electron beam as a localized heat source; (b) SEM images of the two suspended Pt heat flux sensors.

Last but not least, we have tried two approaches to fabricate ultrafine nanostructures on monolayer graphene for the study of wave-like behaviors of heat. We first tested how to introduce nanoscale defects in graphene with a nano-probe and measured the temperature distribution with scanning thermal microscopy (SThM) at the same time. We adjusted the optimal parameters of the probe scanning for the scratching on graphene and introduced nano-to-micro-sized holes on graphene, which exhibited hotspots in the SThM-based temperature mapping.

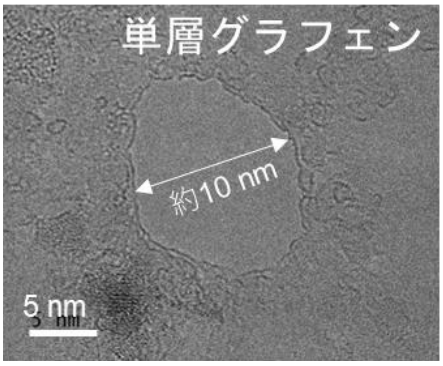

Fig. 3 A hole with a diameter of about 10 nm drilled by focused electron beam

Then, we tested how to drill ultrafine holes in graphene with a focused electron beam. As shown in Fig. 3, with optimized electron beam parameters in a transmission electron microscope, we have fabricated nanoholes with diameters of about 10 nm, which could be hardly realized by other fabrication techniques like the focused ion beam. This kind of ultrafine graphene nanostructures can be utilized to study the wave-like behaviors of heat.

In summary, throughout this project, we have developed high-resolution thermal property measurement methods based on both Raman spectroscopy and scanning electron microscopy. We have also fabricated ultrafine nanostructures in graphene for the study of wave behaviors of phonons.

5．主な発表論文等

〔雑誌論文〕　計12件（うち査読付論文　8件／うち国際共著　2件／うちオープンアクセス　4件）

1．著者名 Li Dawei、Li Qin-Yi、Takahashi Koji	4．巻 198
2．論文標題 Thermal resistance mapping along a single cup-stacked carbon nanotube with focused electron beam heating	5．発行年 2022年
3．雑誌名 International Journal of Heat and Mass Transfer	6．最初と最後の頁 123418～123418
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.ijheatmasstransfer.2022.123418	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

1．著者名 Abe Ryo、Sekimoto Yuki、Saini Shirkant、Miyazaki Koji、Li Qinyi、Li Dawei、Takahashi Koji、Yagi Takashi、Nakamura Masakazu	4．巻 31
2．論文標題 Round Robin Study on the Thermal Conductivity/Diffusivity of a Gold Wire with a Diameter of 30 μm Tested via Five Measurement Methods	5．発行年 2022年
3．雑誌名 Journal of Thermal Science	6．最初と最後の頁 1037～1051
掲載論文のDOI（デジタルオブジェクト識別子） 10.1007/s11630-022-1594-9	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

1．著者名 Li Dawei、Li Qin-Yi、Ikuta Tatsuya、Takahashi Koji	4．巻 120
2．論文標題 Concurrent thermal conductivity measurement and internal structure observation of individual one-dimensional materials using scanning transmission electron microscopy	5．発行年 2022年
3．雑誌名 Applied Physics Letters	6．最初と最後の頁 043104～043104
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1．著者名 Wu Si、Li Qin-Yi、Ikuta Tatsuya、Morishita Kazuhiko、Takahashi Koji、Wang Ruzhu、Li Tingxian	4．巻 171
2．論文標題 Thermal conductivity measurement of an individual millimeter-long expanded graphite ribbon using a variable-length T-type method	5．発行年 2021年
3．雑誌名 International Journal of Heat and Mass Transfer	6．最初と最後の頁 121115～121115
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オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1．著者名	4．巻
Li Qin-Yi、Hao Qing、Zhu Tianhui、Zebarjadi Mona、Takahashi Koji	13
2．論文標題	**5．発行年**
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3．雑誌名	**6．最初と最後の頁**
Engineered Science	24-50
掲載論文のDOI（デジタルオブジェクト識別子）	**査読の有無**
10.30919/es8d1136	有
オープンアクセス	**国際共著**
オープンアクセスとしている（また、その予定である）	該当する

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2022年

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1．発表者名
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2．発表標題
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3．学会等名
第 59 回日本伝熱シンポジウムプログラム
4．発表年
2022年

１．発表者名 Li Qin-Yi
２．発表標題 Controlling thermal conductivity of individual nanocarbon materials
３．学会等名 2021 International Forum on High-end Equipment and Intelligent Manufacturing（招待講演）（国際学会）
４．発表年 2021年

１．発表者名 李秦宜，高橋厚史
２．発表標題 ラマン分光法によるマイクロファイバー同士の接触熱抵抗計測
３．学会等名 第58回日本伝熱シンポジウム
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３．学会等名 International Symposium on Numerical Methods in Heat and Mass Transfer 2020（招待講演）（国際学会）
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４．発表年 2020年

１．発表者名 Li Qin-Yi, Takahashi Koji
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３．学会等名 The 59th Fullerenes-Nanotubes-Graphene General Symposium
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〔図書〕　計0件

〔産業財産権〕

〔その他〕

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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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７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関			
米国	The University of Arizona			