公開

研究終了報告書

「レイフォノニクスによる高度な熱流マネジメント」

研究期間：2019年9月～2023年3月
研究者：Anufriev Roman

1. 研究のねらい

In this project, I aimed to study ballistic phonon transport, its occurrence and limitations, and all the new possibilities available with the concept called ray-phononics. The first goal was to research the fundamentals of ballistic phonon transport and experimentally measure the phonon mean free path in different nanostructures to find optimal range of lengths scales and temperatures for ballistic phonon transport. Next goal was developing the Monte Carlo code and use it to design various phononic nanostructures that use the ballistic phonon transport to shape thermal fluxes. For example, devices that can gradually turn the heat flux direction and ultimately guide the fluxes around an object, thus creating thermal shielding and collimation effects. Next goal was to develop the shifted-probe TDTR method which enables the measurements of heat fluxes from one point to another. Finally, using this setup, I planned to demonstrate some ray-phononic concepts experimentally. The general aim of this project was to investigate the concept of ray-phononics, based on ballistic phonon transport, determine the range of temperatures and distances of ballistic phonon transfer effects, and demonstrate via simulations and new experimental method possible applications, such as thermal guiding, shielding, focusing, and cloaking.

2. 研究成果

(1)概要

In the first year of the project, I focused my efforts on the experimental measurements of the phonon mean free path - the average distance that phonons can travel until a scattering event. I fabricated sets of samples that consist of suspended Si membranes with arrays of nanoslits. The nanoslits had varied spacing between them, which allowed probing the mean free path. Using optical experiments, I measured the thermal conductivity of the samples. In collaboration with French researcher Dr. Jose Ordonez-Miranda, we adopted an analytical model from literature and used it to extract the mean free path spectra of the Si membranes from the measured values of the thermal conductivity.

Our results show that in 145-nm-thin membranes at room temperature phonons can travel ballistically as far as 30 - 300 nm. As we decreased the temperatures, the mean free paths become longer, and the range shifted to 100 - 600 nm at 4 K. These data explain why room-temperature heat conduction in Si appears diffusive at the length scales of several hundreds of nanometers, whereas low-temperature heat conduction at this scale becomes quasi-ballistic. These results have been published in Physical Review B [1].

During the second year, overshadowed by pandemics and inability to continue some of the experimental activities, I focused my efforts on the theoretical part of the project. I performed simulations to study the fundamentals of the ray-phononic concept as well as some devices possible within this paradigm. My simulations demonstrated the formation of directional thermal fluxes in phononic crystals due to the stochastic phonon motion in phononic crystals. I illustrated possible applications of such directional fluxes for emitting directional heat rays, filtering the phonon spectrum, and even protecting a specific region from a thermal gradient. Finally, the simulations showed that this concept is not bound to only low temperatures, and ray-phononic nanostructures can control heat fluxes even at room temperature using modern materials like BAs. Moreover, my simulations suggested that ray phononics is free from limitations of the traditional thermal phononics and enables creating and guiding thermal fluxes in realistic nanostructures regardless of their surface roughness. Thus, my work this year established theoretical grounds for this project and demonstrated potential applications of the project. These results have been published in Materials Today Physics [2] and Applied Physics Letters.

During the third fiscal year, I focused my efforts on switching the project material from Si to SiC because Si showed short phonon mean free path during the first year of the project. Thus, I developed the fabrication process on the SiC membranes, which was complicated by the fact that SiC is a more durable material than Si and resists traditional plasma etching. However, the fabrication process has been adapted to produce nanoscale features on SiC, and I could fabricate various silicon carbide nanostructures.

Next, I measured the obtained membranes, nanowires, and phononic crystals to better understand the nanoscale thermal properties of SiC, which have remained mostly unknown. I demonstrated how the thermal conductivity scales down with the limiting dimension of the structure and be as low as 35 W/m·K in nanostructures with the limiting dimension of 30 nm, which contrasts with bulk values of more than 300 W/m·K. Moreover, I measured the phonon mean free path in SiC membranes and found that it is longer than that in Si, which is beneficial for the project goals. These results have been published in NPG Asia Materials [3].

In the last year, I developed the shifted-probe time-domain thermoreflectance method (TDTR), fabricated the setup, samples, and, in collaboration with another French researcher Dr. Laurent Jalabert, developed the software for the setup. Moreover, with a student, we simulated more ray-phononic devices using the Monte Carlo code and proposed a thermal collimator based on a parabolic thermal mirror. We attempted to measure this and other effects experimentally, using the developed shifted probe setup. During the experiments, we observed that the sensitivity of the experiment is likely insufficient to probe the expected effects. Thus, further optimization of the setup and the sample is presently performed.

(2)詳細

During the first fiscal year, I focused my efforts on the experimental measurements of the phonon mean free path. First, I fabricated two sets of samples consisting of suspended silicon membranes with arrays of nanoslits, shown in Fig. 1. The nanoslits had varied spacing (*w*) between them, which allowed probing the mean free path. Moreover, to ensure that our method does not depend on a particular design of the pattern, we fabricated two sets of samples with periodicity (*p*) of 500 and 1000 nm. Using the TDTR techniques, I measured the thermal decay time characteristics of all samples at different temperatures. Then, using the Finite Element analysis, I converted the measured data into the thermal conductivity for both sample sets.

Next, in collaboration with French researcher Dr. Jose Ordonez-Miranda, we adopted the semi-analytical model from the literature and extracted the mean free path spectra of the silicon membranes from the measured values of the thermal conductivity. Figure 1c shows the obtained mean free path accumulation functions at different temperatures and theoretical predictions in the literature.

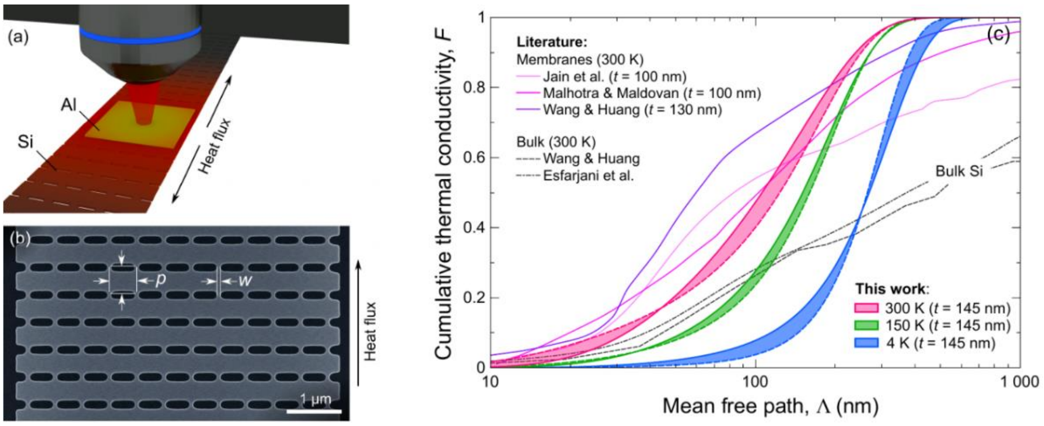

Figure 1. (a) Scheme of the TDTR experiments and (b) SEM image of Si sample membrane with array of slits. (c) Measured mean free path spectra in a 145-nm-thick Si membrane vs. theoretical prediction in the literature.

The results show that the mean free path in 145-nm-thin Si membranes spans from 30 to 300 nm at room temperature. As we decreased the temperatures, the mean free paths become longer, and the range shifted to 100 – 600 nm at 4 K. These data explain why room-temperature heat conduction appears diffusive at the length scales of several hundreds of nanometers, whereas low-temperature heat conduction at this scale becomes quasi-ballistic. These results have been published in Physical Review B [1].

During the second fiscal year, overshadowed by the quarantine, I focused my efforts on theoretical side of the project and simulated the devices based on the principles of ray phononics. To simulate the phonon transport under particle approximation I have implemented a python code of a Monte Carlo algorithm. Figure 2a illustrates the simulated thin silicon membranes with arrays of holes. Phonons are traced inside these structures as particles traveling in straight lines. The algorithm traces trajectories of tens of thousands of phonons and builds thermal energy maps.

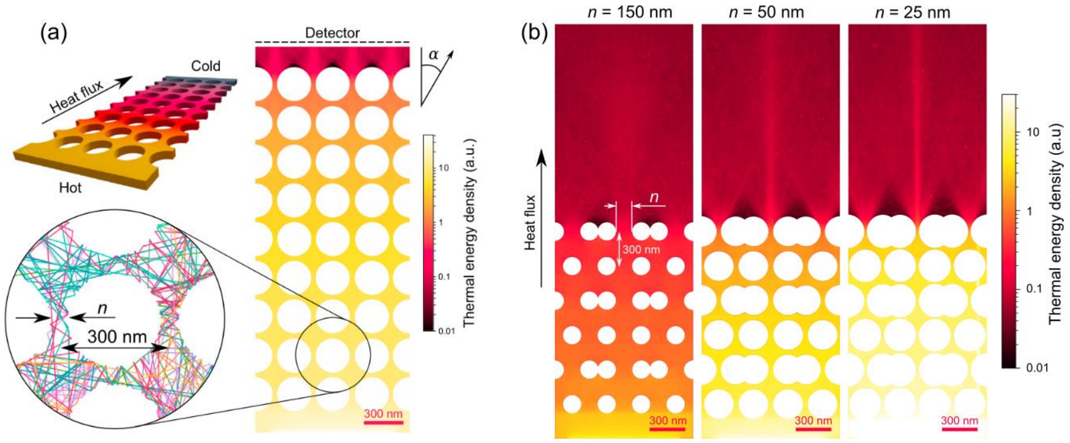

Figure 2. (a) Illustration of a simulated phononic structure with an example of its thermal energy map. The inset shows the phonon trajectories. (b) Thermal energy maps of the Si phononic structures that act as a source of directional heat flux 4 K. The emitted heat ray becomes stronger as the channel narrows.

Simulations demonstrated that arrays of holes can shape overall heat flux and create directional phonon fluxes in the passages between the holes. This effect lies in the core of the ray phononic concept. My simulations helped to better understand this effect and its cause. Simulations also demonstrated that the strength of this effect depends on the number of hole periods in the structure, inter hole distance, and hole shapes. Next, I demonstrated how the directional fluxes can be emitted as heat ray in solid material. Figure 2b shows the simulation of a structure emitting a heat ray from the middle passage whereas other passages are blocked. As the passage narrows, the heat ray becomes stronger. To illustrate a possibility of application, I also demonstrated a thermoelectric device based on this principle.

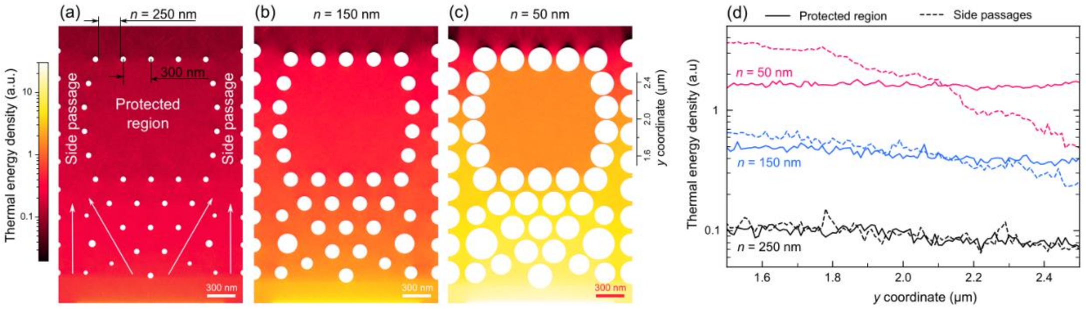

Figure 3. (a-c) Thermal energy maps of the structures with n = 250, 150, and 50 nm show how the energy in the protected region plateaus as the channels narrow. (d) Profiles of thermal energy density measured in the protected region and along the side channels also show that in the structure with n = 50 nm the protected region feels no thermal gradient.

Another example of how ray phonics can be applied is the structure protecting a region from a thermal gradient. Figure 3a-c shows the design of the nanostructure, in which the hole pattern guides the heat around the protected region. To demonstrate the protection from thermal gradient, I simulated three structures with different widths of the channels (n) and observed changes in the thermal gradients. The energy maps of all three structures (Figs. 3a-c) show that the thermal gradient continues in the channels around the protected region. But inside the protected region, the thermal energy gradually becomes uniform and forms a plateau as the channels narrow. Moreover, I have also estimated the length and temperature scales at which the effects can be experimentally observable. These results motivated me to switch my fabrication process to SiC instead of Si. These results have been published in Materials Today Physics [2].

Since my research in the first fiscal years demonstrated that Si has a short phonon mean free path, I decided to switch the material to SiC, which is known for its long mean free path. Thus, most of my efforts in the third fiscal year have been devoted to the development of fabrication methods that would enable fabrication of silicon carbide nanostructures on suspended 3C SiC membranes. The issues with this fabrication approach included extreme fragility of thin nanomembranes suspended over 1 × 1 mm and high resistance of silicon carbide to the standard dry etching process. However, I resolved these issues and successfully fabricated silicon carbide nanostructures.

Then, I measured the obtained samples using TDTR method and retrieved the thermal conductivity of silicon carbide nanomembranes, nanowires, and phononic structures. Remarkably, the thermal conductivity of nanostructures scales down proportionally to the narrowest dimension of the structure. For example, for the smallest dimension of 60 nm, the thermal conductivity can be as low as 35 W/m·K, an order of magnitude lower than the values reported for bulk silicon carbide (> 300 W/m·K). Figure 4 summarizes the obtained data and shows that the points form a trend as a function of the limiting dimensions.

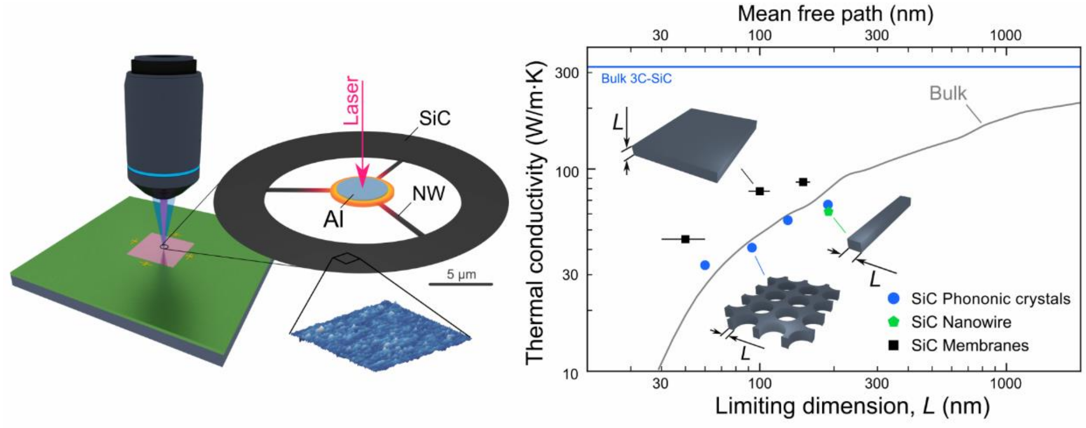

Figure 4. Scheme of TDTR experiment on SiC nanostructures and measured thermal conductivity of various SiC nanostructures vs their limiting dimensions and bulk mean free path.

In addition, I also measured the phonon mean free path in SiC membranes to ensure that this material is superior to Si. Indeed, the measured mean free path spectrum of silicon carbide was longer than that in silicon. However, at low temperatures, the mean free path spectra of Si and SiC are nearly identical due to the absence of phonon-phonon scattering processes. Thus, SiC provides a sufficient advantage over Si only at high temperatures. These results have been published in NPG

Asia Materials [3].

In the last year of the project, we developed the shifted-probe TDTR method, assembled the setup, samples, and, in collaboration with another French researcher Dr. Laurent Jalabert, developed the software for the setup. The scheme of the setup and a membrane sample are shown in Fig. 5. The proof-of-concept experiment shows that as the distance between pump and probe pad is increased, the delay in the signal is also increased proportionally (Fig. 5c). This result demonstrates that the setup is functioning as expected and is implemented successfully. To process the data from this experiment I developed software that fits delayed peak and compares it with the prediction of the finite element model. The value of the thermal conductivity obtained with this new method agree within 10% uncertainty with values extracted by traditional TDTR method on the same sample illustrated in Fig. 5. Thus, this method can become an alternative to the traditional TDTR method.

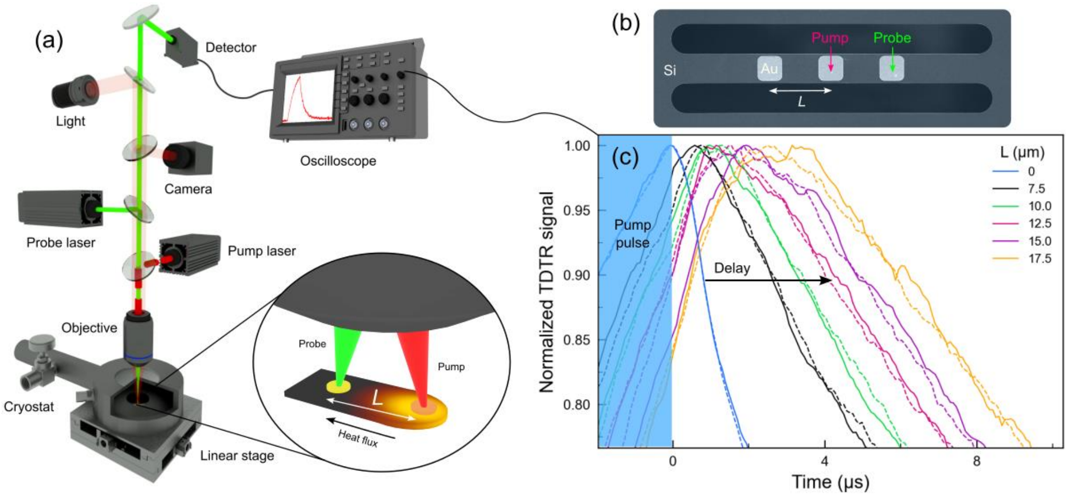

Figure 5. (a) Scheme of the shifted-probe TDTR setup. (b) Sample with varied distance between pump and the probe. (c) TDTR signal show the delay proportional to the distance between the pump and the probe.

Moreover, with a student, we simulated more ray-phononic devices using the Monte-Carlo code and proposed a thermal collimator based on a parabolic thermal mirror . We attempted to measure this effect experimentally, using the developed shifted probe setup. During the experiments, we observed that the sensitivity of the experiment is likely insufficient to probe the expected effects.

Finally, I also tried to measure the ray-phononic effect of creating directional beams illustrated in Fig. 2. The idea of the experiment was to compare the difference in the delay between the peaks measured on aligned and staggered lattice of holes. While the result was generally positive and the difference was observed, the measured difference was comparable to the uncertainty of the measurement. Thus, further optimization of the setup and the sample design is taking place at the time of writing.

3. 今後の展開

Based on the current experimental results, it appears that the shifted-probe TDTR experiment lacks sensitivity because the width of the shifted peak is comparable to the shift. Thus, the sample and the setup shall be optimized to produce narrower peaks with larger shifts. This sensitivity can be optimized by reducing the size of the probed spot and by placing the probe spot closer to the heat sink. Moreover, implementation of the balanced photodetector should improve the sensitivity of the system. In any case, the shifted probe setup already proved to be a useful metrology tool. For example, it already found application for simplified measurements of thermal radiation from a hot to cold body. Typically, such experiments require complex sample fabrication and measurements, involving Joule heating and sensing via change in resistance. Using the shifted-probe TDTR setup and SiC nanostructures fabricated on a suspended wafer via methods developed in this project, I was able to measure thermal radiation from a hot to cold plate in the near-field regime. Thus, this contactless experimental method is expected to facilitate thermal measurements at nanoscale and may replace more complex electrical methods in a few years.

Another area that could be improved is the nanofabrication on SiC membranes. In general, SiC is regarded as a material for power microelectronics. However, the high mechanical resistance of this material makes the nanofabrication exceptionally difficult. During this project I made certain progress which enabled me to fabricate SiC nanostructures such as SiC nanowires and phononic crystals. However, the quality of these nanostructures is still not as perfect as their analogues made of Si. Thus, to fully establish SiC as a viable option for microelectronics, further effort should be made to improve the fabrication precision and quality. This can take from a few months to a year. Moreover, this project showed that the thermal conductivity of SiC nanostructure is nowhere near the bulk values often cited in the literature. Thus, this project equipped engineers with more realistic estimates for thermal performance of SiC nanostructures.

On the theoretical part of the project, the development of the Monte Carlo code has been mostly completed, the code has been open-sourced, and now is available to researchers and engineers interested in nanoscale thermal transport simulations and ray-phononic devices. Future developments of the code may include parallelization of the computation to improve performance and may take a year.

4. 自己評価

Overall, I find that most of the goals of the project have been achieved. Specifically, the technique to measure and analyze the mean free path spectra has been developed in collaboration with French researcher Dr. Jose Ordonez-Miranda. With this method, we measured ballistic phonon spectra for Si and SiC nanomembranes at various temperatures. This technique can now be used to probe other materials and nanostructures, which should help researchers and engineers to better understand thermal properties of microelectronic devices.

On the theoretical side of the project, I successfully developed the Monte Carlo code and conducted all the planned simulations. Now this code is open-sourced under the name “FreePATHS” and can be used by researchers all over the world to easily assess thermal performance of nanostructures with complex geometries.

On the fabrication front, we developed the nanofabrication process on crystalline SiC, which enables further advance and miniaturization of power microelectronic devices. The measured and reported thermal properties of SiC will certainly help the industry to design better SiC nanoscale devices.

Finally, in collaboration with another French researcher Dr. Laurent Jalabert, we successfully implemented the novel shifted-probe TDTR method. Although early experiments showed that the sensitivity of this method can be improved, in principle the method functions as intended and has already found application in the measurements of thermal radiation, measurements of surface polariton propagation, and more common thermal measurements.

However, the experimental demonstration of ray-phononic devices using shifted-probe TDTR method has only started, and the experiments were not yet performed to the extent they were planned to, at least at the time of writing. It became clear from the early experiments that the method requires better design of the samples and impartments in sensitivity. Moreover, the experimental demonstration using SNOM method, in collaboration with a PRESTO researcher Prof. Kajihara, has not yet been completed. Although we optimized the sample design after several trials, some difficulties seem to remain on the measurement side. Thus, this part of the project has not been completed to the full extent yet.

I general, the project has impacted and advanced various areas of thermal science, by providing new fundamental knowledge of the phonon properties, producing open-source simulation tools, finding new fabrication techniques for SiC nano-structuring, and introducing a new measurement method. It is my hope that these innovations will help researchers in Japan and all over the world to advance towards more efficient and environmentally friendly microelectronics.

5. 主な研究成果リスト

（1）代表的な論文（原著論文）発表

研究期間累積件数：9件

1. Measurement of the phonon mean free path spectrum in silicon membranes at different temperatures using arrays of nanoslits, Physical Review B 101 (11), 115301 (2020)

In this work, we experimentally probe the phonon MFP spectra in suspended silicon membranes. First, we measure the thermal conductivity of membranes with arrays of slits at different temperatures. Next, we develop a fully analytical procedure to extract the accumulated thermal conductivity as a function of the MFP. The measured phonon MFP in 145-nm-thick membranes with the surface roughness of 0.2 nm is shorter than that in bulk due to the scattering at the membrane boundaries. At room temperature, the phonon MFP does not exceed 400 nm. However, at 4 K, the MFP becomes longer, and some phonons can travel ballistically for up to one micrometer. These results thus shed light on the long-lasting question of the range of ballistic phonon transport at different temperatures in nanostructures based on silicon membranes.

2. Ray phononics: thermal guides, emitters, filters, and shields powered by ballistic phonon transport, Materials Today Physics 15, 100272 (2020)

In this paper, we propose an alternative concept of heat manipulation based on the particle picture of phonons and their ballistic transport. We call this concept ray phononics as a thermal analogy of ray optics. Ray phononics is free from limitations of the traditional phononics and enables creating and guiding thermal fluxes in realistic nanostructures regardless of their surface roughness. Our simulations illustrate possible applications of ray phononics for emitting directional heat rays, filtering the phonon spectrum, and even protecting a specific region from a thermal gradient. Finally, we show that the proposed concept is not bound to only low temperatures, and ray-phononic nanostructures can control heat fluxes even at room temperature using modern materials like boron arsenide.
3. Nanoscale limit of the thermal conductivity in crystalline silicon carbide membranes, nanowires, and phononic crystals, NPG Asia Materials 14, 35 (2022)
In this work, we systematically study heat conduction in SiC nanostructures, including nanomembranes, nanowires, and phononic crystals. Our measurements show that the thermal conductivity of nanostructures is several times lower than that in bulk and that the values scale proportionally to the narrowest dimension of the structures. In the smallest nanostructures, the thermal conductivity reached 10% of that in bulk. To better understand nanoscale thermal transport in SiC, we also probed phonon mean free path and coherent heat conduction in the nanostructures. Our theoretical model links the observed suppression of heat conduction with the surface phonon scattering, which limits the phonon mean free path and thus reduces thermal conductivity. This work uncovers thermal characteristics of SiC nanostructures and explains their origin, thus enabling realistic thermal engineering in SiC microelectronics.

（2）特許出願

研究期間全出願件数：0 件（特許公開前のものも含む）

（3）その他の成果（主要な学会発表、受賞、著作物、プレスリリース等）

- (invited) R. Anufriev et al., “Ballistic heat conduction at nanoscale: demonstrations and applications” Telluride workshop: Thermal Transport at the Nanoscale, Telluride, USA (2022).
- R. Anufriev et al., “Nanoscale limit of the thermal conductivity in SiC membranes, nanowires, and phononic crystals”, Nanoscale and Microscale Heat Transfer VII, P.22, Palermo, Italy (2022).
- R. Anufriev, et al., “Phonon mean free path spectroscopy in Si and SiC nanomembranes in the 4 - 400 K range”, MRS Spring Meeting and Exibits , SF15.05.03, Hawaii, USA (2022).
- R. Anufriev and M. Nomura, “Ray Phononics for Advanced Heat Flux Manipulations in Ballistic Regime”, 2021 Virtual MRS Spring Meeting & Exhibit, NM08.06.02, online (2021).
- The Junior Prize of the IPPA (2019)
- Young Scientist Award in Science and Technology by the Ministry of Education, Culture, Sports, Science and Technology (2023)