An Integrated H-type Method to Measure Thermoelectric Properties of Two-Dimensional Materials

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Abstract

This paper presents a new integrated H-type method for precisely characterizing the thermoelectric properties of suspended two-dimensional (2D) materials. The current method combines the micro-device electrical measurement and laser heating together. The electrical measurement offers high accuracy, while the measurement principle and operation can be much simplified by using the noncontact laser heat source. The 2D material is suspended between two metallic nanofilms to form a H-type structure, i.e. named as H-type method. The metallic nanofilms can be used as an electrical Joule heater and a temperature sensor. By simply changing the external circuit, the electrical conductivity, thermal conductivity and Seebeck coefficient can be measured on the same nanomaterial sample, simultaneously. Thus, the main origin of measurement uncertainty caused by the sample discrepancy can be avoided. In the measurement, the laser absorption rate of the 2D material can be obtained as well. Taking monolayer graphene as an example, a detailed uncertainty analysis was carried out. This work provides a reliable and accurate measurement method to achieve full thermoelectric properties of 2D materials, setting a foundation for practical design of efficient 2D thermoelectric devices.

Keywords: 2D materials; Thermoelectric properties; H-type method; Non-contact laser heating.

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1. Introduction

The rapid increase in global demand for clean energy has led to a strong interest in thermoelectric materials that can convert thermal energy into electrical energy.^[1-3] However, due to the low energy conversion efficiency of thermoelectric materials, their applications are still limited.^[4] The key parameter of thermoelectric materials is the figure of merit $ZT = S^2 T \sigma / \lambda$, where S is Seebeck coefficient, σ is electrical conductivity and λ is thermal conductivity, respectively. In recent years, it has been found that a higher ZT can be obtained in low-dimensional materials, such as super-lattices structures,^[5] nanocomposites^[6] and quantum dots.^[7] Among the low-dimensional materials, the two-dimensional (2D) materials, such as graphene, black phosphorus, etc., have attracted much attention. For example, Seol *et al.* measured the three key parameters of supported graphene^[8] and Li *et al.* fabricated as-grown suspended graphene nanoribbons, which yielded ZT value up to 0.1.^[9]

Characterization of the thermoelectric properties of 2D materials is essential to understand the fundamental physical mechanism and development of effective thermoelectric devices. Unfortunately, most of the current methods can only measure these three thermoelectric parameters, i.e. electrical conductivity, thermal conductivity and Seebeck coefficient, separately. For nanoscale materials, the sample discrepancy due to different crystalline structure or preparation conditions may cause significant measurement uncertainty when switching the sample to measure each parameter. The best way is to use the same nanomaterial sample to obtain the three thermoelectric parameters simultaneously. But there are currently very few methods to meet this goal.^[10-13]

In 1958, Harman proposed a method that can directly obtain the figure of merit of the sample.^[10] In the Harman method, a direct current (DC) passes through a homogeneous rod of thermoelectric material longitudinally. Generated Joule heat can be transferred through the Peltier effect or heat conduction at the interface. Under the steady-state adiabatic conditions, the heat transferred by the Peltier effect is equal to the heat transferred by thermal conduction. Under this heat balanced thermal condition, the figure of merit of the

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thermoelectric material can be calculated. However, the thermal radiation and convection have noticeable influences on the final result. Also, it is difficult to use the Harman method to measure the nanomaterials.

In 2001, P. Kim *et al.* proposed a suspended micro-device method to comprehensively measure the thermoelectric properties of nanomaterials.^[11] The test sample was suspended between two micro-pads with platinum wire deposited on top as heater and temperature sensors, respectively. One of the micro-pads is used for heating at a large DC current. The other one is used as resistance thermometer at a small DC current. The temperatures of two micro-pads can be regarded as a stable value during the measurement. In the electrical conductivity measurement, the voltage and current through the sample can be directly measured. In the thermal conductivity measurement, a temperature gradient is built between two micro-pads. The temperature of each pad can be measured separately by monitoring its resistance change. The thermal conductivity of sample can be calculated after measuring the heat flow and temperature difference between two micro-pads. In the Seebeck coefficient measurement, the temperature difference and potential difference between two micro-pads can be measured at the same time, then the Seebeck coefficient of the sample bridged between two micro-pads can be calculated. However, the temperature of the heater is not uniform when the thermal conductivity of the 2D material such as graphene is large, resulting in a relatively large error. Besides the thermal radiation will cause non-negligible influence on the measure result.

T-type method is also a representative method for measuring multiple thermoelectric properties of nanomaterials.^[12] In this method, the nanomaterial is cantilevered with one end attached to the center of a suspended platinum nanofilm as a thermometer and Joule heater simultaneously. The electrical conductivity can be determined by a standard four-probe method. By comparing the average temperature rise of the platinum nanofilm before and after attaching the nanomaterial to the platinum nanofilm, the thermal conductivity can be obtained. However, in the Seebeck coefficient measurement, two sensors that respectively work as a Joule heater and a thermometer are needed. The T-type method had only one sensor and thus it needed to use a complex AC heating/DC detecting scheme.

In this report, a comprehensive H-type method was developed that can simultaneously measure the electrical conductivity, thermal conductivity and Seebeck coefficient of the same 2D nanomaterial in situ. A 2D material sample was suspended between two metallic nanofilms, forming a H-type structure. Two ends of the metallic nanofilms were connected to the electrode pads for electrical measurement. The metallic nanofilm can be used as a resistance temperature sensor and electrical heater at the same time. In the measurement of Seebeck coefficient, a laser beam was focused to heat the 2D material sample at different positions and caused an

asymmetric temperature distribution along the sample. The thermoelectric potential of the sample can be directly measured at two metallic nanofilms. The laser absorption rate is also important for nanomaterials.^[14-16] In H-type method, the laser absorption rate of 2D material can be obtained as well. Compared with T-type method, H-type method has two sensors, and the measurement accuracy and reliability can be much improved. Besides, there is no complicated experimental measurement and data analysis system for measuring Seebeck coefficient. The current method can be performed on different suspended 2D materials and offers a high measurement accuracy.

2. Experimental section

2.1. Measurement system

As shown in Fig. 1, a free-standing 2D material was prepared following a series of Micro-Electro-Mechanical System (MEMS) process discussed in our previous paper.^[17-19] The 2D material sheet was suspended between two metallic nanofilms, named A and B in Fig. 1. The 2D material sample and two sensors form a H-type structure, thus the current method is called H-type method. The nanofilm sensor was 100 nm thick Au film deposited on SiO₂/Si by using a physical vapor deposition method. The geometric parameters of metallic nanofilms A, B and the 2D material were accurately measured by using scanning electron microscope (SEM) imaging. After etching the SiO₂ layer and Si substrate, two sensors were fully suspended. Two large-area electrode pads were deposited at both sides of sensor. Each electrode pad has two contacts for connecting thin lead wires. There are four electrode pads and eight contacts in total. The whole Si chip with MEMS testing device was placed on the temperature control platform, so as to accurately control the environmental temperature at T_0 . The experiment was carried out in a high vacuum chamber (10^{-4} Pa). Both Au nanofilms A and B can be used as electrical heater and electrical resistance thermometer. The resistance temperature coefficient and thermal conductivity of the metallic nanofilms were calibrated in advance. The well calibrated nanofilm sensor can offer a temperature measurement accuracy better than 0.01 K.

2.2. Electrical conductivity measurement

Fig. 2 shows the electrical circuit used for measuring the electrical conductivity of 2D material. The Kelvin four-wire method was used to measure electrical conductivity, thus the resistance of the metal wire in the external circuit can be eliminated. The voltage U_2 and the current I_2 flowing through the 2D material were measured by a high-precision digital multimeter Keithley 2002. The formula for calculating the conductivity of 2D materials is shown in Equation (1):

$$\sigma = \frac{I_2 a_2}{L_2 d_2 U_2} \quad (1)$$

where U_2 is the voltage of the 2D material and I_2 is the current flowing through the 2D material. L_2 , a_2 , d_2 are the length, width and thickness of the 2D material, respectively.

Fig. 1 Schematic diagram of H-type structure. The 2D material is suspended between two metallic nanofilms A and B. The metallic nanofilm sensor is connected with two electrode pads at both sides. Each electrode pad has two contacts for connecting thin lead wires. There are four electrode pads and eight contacts in total. All the metallic nanofilm and pads were made of 100 nm thick Au film by using a physical vapor deposition method.

Fig. 2 Schematic diagram of electrical conductivity measurement. V_2 (connecting contact 2 and contact 4) and A_2 (connecting contact 1 and contact 3) are the high-precision digital multimeters used to measure the voltage and current of the 2D material.

2.3. Thermal conductivity measurement

Fig. 3 Schematic diagram of thermal conductivity measurement. V_1 , A_1 and V_2 , A_2 are high-precision digital multimeters used to measure the voltage and current of metallic nanofilms A and B.

Fig. 3 shows the electrical circuit used for measuring the thermal conductivity of 2D material. A larger electrical heating power was applied on the metallic nanofilm A, thus a significant temperature rise was built on A. A small electrical power was applied on the metallic nanofilm B to measure its resistance.

The voltages U_1 , U_3 , currents I_1 , I_3 and $R_{\lambda 1}$, $R_{\lambda 3}$ of the metallic nanofilms A, B were measured in the experiment. The average temperatures $T_{\lambda 1}$, $T_{\lambda 3}$ of two metallic nanofilms A and B were calculated through the linear relationship between temperature and resistance.

Fig. 4 shows the 2D heat conduction model to calculate thermal conductivity, where the midpoint of the metallic nanofilm A is selected as the zero-point of the x-axis coordinate along the metallic nanofilm A, and the y-axis direction is perpendicular to the nanofilm A. The governing equations of heat conduction in metallic nanofilms A, B and 2D material are given as follows:

$$\left\{ \begin{array}{l} \text{metallic nanofilm A : } \left\{ \begin{array}{l} \frac{d^2 T_1(x)}{dx^2} + \frac{U_1 I_1}{L_1 a_1 d_1 \lambda_1} - \frac{T_1(x) - T_2(x, 0)}{d_1 \lambda_1 R_C} = 0, 0 \leq x \leq \frac{L_2}{2} \\ \frac{d^2 T_1(x)}{dx^2} + \frac{U_1 I_1}{L_1 a_1 d_1 \lambda_1} = 0, \frac{L_2}{2} < x \leq \frac{L_1}{2} \end{array} \right. \\ \text{2D material : } \frac{\partial^2 T_2(x, y)}{\partial x^2} + \frac{\partial^2 T_2(x, y)}{\partial y^2} = 0 \\ \text{metallic nanofilm B : } \left\{ \begin{array}{l} \frac{d^2 T_3(x)}{dx^2} - \frac{T_3(x) - T_2(x, a_2)}{d_3 \lambda_3 R_C} = 0, 0 \leq x \leq \frac{L_2}{2} \\ \frac{d^2 T_3(x)}{dx^2} = 0, \frac{L_2}{2} < x \leq \frac{L_3}{2} \end{array} \right. \end{array} \right. \quad (2)$$

where $T_1(x)$, $T_2(x, y)$ and $T_3(x)$ are the temperature distribution of metallic nanofilm A, 2D material and metallic nanofilm B, respectively. λ_1 and λ_3 are the thermal conductivity of metallic nanofilms A and B. R_C is the contact thermal resistance between the 2D material and the metallic nanofilm. In this work, the metallic nanofilm was deposited on 2D material as the temperature sensor and the contact thermal resistance is significantly smaller than that made by transferring 2D material onto the metallic sensor with air gap or contaminations at the interface.^[20,21]

The boundary condition at $x=0$ is:

$$\left. \frac{dT_1(x)}{dx} \right|_{x=0} = \left. \frac{\partial T_2(x, y)}{\partial x} \right|_{x=0} = \left. \frac{dT_3(x)}{dx} \right|_{x=0} = 0 \quad (3)$$

The boundary conditions at $x=L_2/2$ are:

$$\left. \frac{\partial T_2(x, y)}{\partial x} \right|_{x=L_2/2} = 0 \quad (4)$$

The boundary conditions at $x=L_1/2$ and $x=L_3/2$ are:

$$T_1(L_1/2) = T_3(L_3/2) = T_0 \quad (5)$$

The mathematical derivation process for analytical solution of the above equations is quite complicated, which is given in supplementary material. The thermal conductivity of 2D material can be expressed by Equation (6):

$$\lambda_2 = - \frac{T_1(x) - T_2(x, 0)}{R_C \left. \frac{\partial T_2}{\partial y} \right|_{y=0}} \quad (6)$$

The analytical solution of temperature distribution in Fig. 4 is complicated and difficult to use directly. Also, there is no analytical solution available for complicated geometric shape of 2D material rather than rectangle. In the following analysis, a numerical solution was used instead to obtain the thermal conductivity λ_2 of the 2D material. The accuracy of numerical simulation has been proved by the analytical solution in advance. An iterative calculation process is needed to obtain the thermal conductivity of 2D material sample as follows:

Step 1: Given an initial thermal conductivity λ_2 of 2D material based on the literature value. The selection of the initial thermal conductivity does not affect the final result, but the process of iterative convergence.

Step 2: Perform numerical simulation to obtain the entire temperature distribution of nanofilm sensors and the 2D

materials and then calculate the average temperatures $T_{\lambda A}$, $T_{\lambda B}$ of the metallic nanofilms A and B.

Step 3: Compare $T_{\lambda A}$ and $T_{\lambda B}$ obtained by numerical simulation in step 2 with $T_{\lambda 1}$ and $T_{\lambda 3}$ measured in the experiment. There are three comparison results:

Result 1: If $|T_{\lambda A} - T_{\lambda 1}| < 0.01$ K, and $|T_{\lambda B} - T_{\lambda 3}| < 0.01$ K, it means that the predetermined thermal conductivity in the numerical simulation is the real thermal conductivity.

Result 2: If $T_{\lambda A} > T_{\lambda 1}$ and $T_{\lambda B} < T_{\lambda 3}$, then it means that the predetermined thermal conductivity in the numerical simulation is too small. The heat conducted from the metallic nanofilm A to B is too small, resulting in larger temperature of difference between $T_{\lambda A}$ and $T_{\lambda B}$. Thus, the initial thermal conductivity in calculation should be increased and repeat step 2.

Result 3: If $T_{\lambda A} < T_{\lambda 1}$ and $T_{\lambda B} > T_{\lambda 3}$, then it means that the predetermined thermal conductivity in the numerical simulation is too large. Too much heat is conducted from the metallic nanofilm A to B, resulting in smaller temperature difference between $T_{\lambda A}$ and $T_{\lambda B}$. Thus, the initial thermal conductivity in calculation should be decreased and repeat step 2.

After repeating the above iteration processes, the true thermal conductivity of 2D material can be decided when the convergence Result 1 is satisfied.

Fig. 4 Schematic diagram of mathematical model to calculate thermal conductivity. The midpoint of the metallic nanofilm A is selected as zero-point of the x-axis coordinate along the metallic nanofilm. The y-axis is perpendicular to the metallic nanofilm A. L_1 , a_1 and L_3 , a_3 are the length, width and thickness of the metallic nanofilm A and B, respectively. L_2 , a_2 are the length and width of the 2D material.

2.4. Seebeck coefficient measurement

Fig. 5 Schematic diagram of Seebeck coefficient measurement. A laser beam was focused on one side of the center point of the 2D material to produce an asymmetric temperature distribution. V_1 , A_1 and V_3 , A_3 are high-precision digital multimeters used to measure the voltage and current of the metallic nanofilms A and B. V_2 is used to measure the potential difference across the 2D material.

The Seebeck coefficient is defined as the potential difference divided by the temperature difference. Fig. 5 shows the electrical circuit used for measuring the Seebeck coefficient of 2D material. A laser beam was focused on one side of the center point of the 2D material to produce an asymmetric temperature distribution. The laser spot radius is r and the heating power is q . Both metallic nanofilms A and B served as electrical resistance thermometer for 2D material. The temperature rise of metallic nanofilm caused by Joule heating is negligible. T_{S1} , and T_{S3} are the measured average temperatures of metallic nanofilms A and B, respectively. The potential difference U_2 across 2D material caused by asymmetric heating was measured as well.

Fig. 6 shows the mathematical model of heat conduction with laser heating. The governing equations (7) of metallic nanofilms A, B and the 2D material are given as follows:

$$\left\{ \begin{array}{l} \text{metallic nanofilm A:} \left\{ \begin{array}{l} \frac{d^2 T_1(x)}{dx^2} - \frac{T_1(x) - T_2(x, 0)}{d_1 \lambda_1 R_c} = 0, 0 \leq x \leq \frac{L_2}{2} \\ \frac{d^2 T_1(x)}{dx^2} = 0, \frac{L_2}{2} < x \leq \frac{L_1}{2} \end{array} \right. \\ \text{2D material:} \quad \frac{\partial^2 T_2(x, y)}{\partial x^2} + \frac{\partial^2 T_2(x, y)}{\partial y^2} + \frac{\eta q}{\pi r^2 d_2 \lambda_2} \exp(-(x^2 + (y - c)^2) / r^2) = 0 \\ \text{metallic nanofilm B:} \left\{ \begin{array}{l} \frac{d^2 T_3(x)}{dx^2} - \frac{T_3(x) - T_2(x, a_2)}{d_3 \lambda_3 R_c} = 0, 0 \leq x \leq \frac{L_2}{2} \\ \frac{d^2 T_3(x)}{dx^2} = 0, \frac{L_2}{2} < x \leq \frac{L_3}{2} \end{array} \right. \end{array} \right. \quad (7)$$

where, r , q , and c are the laser radius, power and focus position, respectively. η is the laser absorption rate of the 2D material. The boundary conditions are the same as the Eq. (2).

Fig. 6 Schematic diagram of mathematical model to calculate Seebeck coefficient. Laser heating is used to create an asymmetric temperature distribution, where c , r are the focus position and the laser radius, respectively.

Due to the complicated heat source term, the analytical solution cannot be found and the numerical simulation was used to calculate the temperature distribution. Here the unknown laser absorption rate η can be determined as well. The calculation process is given as follows:

Step 1: An initial value of the laser absorption rate was set to be 0.05, and then the whole temperature distribution in Fig.6 can be obtained by numerical simulation.

Step 2: According to the simulation results, the average temperatures T_{SA} and T_{SB} of the metallic nanofilms A and B were calculated respectively.

Step 3: Compare T_{SA} and T_{SB} obtained by numerical simulation in step 2 with T_{S1} and T_{S3} measured in the experiment. There are three results:

Result 1: If $|T_{SA}-T_{S1}|<0.01K$ and $|T_{SB}-T_{S3}|<0.01K$, it means that the laser absorption rate η given in the numerical simulation is true.

Result 2: If $T_{SA}>T_{S1}$ and $T_{SB}>T_{S3}$, it means that the laser absorption rate given in the numerical simulation is too large, resulting in higher temperature of metallic nanofilm than the experimental value. Then, the laser absorption rate η should be decreased and repeat step 1.

Result 3: If $T_{SA}<T_{S1}$ and $T_{SB}<T_{S3}$, it means that the laser absorption rate η given in the numerical simulation is too small, resulting in smaller temperature of metallic nanofilm than the experimental value. Then, the laser absorption rate η should be increased and repeat step 1.

After repeating the above iteration processes, the true laser absorption rate of the 2D material can be decided when

the convergence Result 1 is satisfied. Thus, the true temperature distribution of the whole nanofilm sensor and 2D material under laser heating can be obtained. So, the temperature difference $T_{2A}-T_{2B}$ at both ends of the 2D material can be calculated and the Seebeck coefficient can be determined as:

$$S = \frac{U_2}{T_{2A} - T_{2B}} \quad (8)$$

where T_{2A} is the average temperature of 2D material at end A, and T_{2B} is the average temperature of 2D material at end B. U_2 is the potential difference across the 2D material.

3. Simulation and uncertainty analysis

Graphene is currently the mostly used 2D material, and there have been many reports on the properties of graphene.^[22-24] Here, we take graphene as an example to demonstrate the feasibility of current method and analyze its uncertainty. The recommended graphene properties in literature are used here: the electrical conductivity is $\sigma=4\times10^5\Omega^{-1}m^{-1}$,^[24] the thermal conductivity^[23] is $\lambda_2=2400Wm^{-1}K^{-1}$ and the Seebeck coefficient^[22] is $S=70\mu VK^{-1}$. The metallic nanofilms were made of gold, whose thermal conductivity is $\lambda_1=\lambda_3=317Wm^{-1}K^{-1}$. The geometric dimensions are given as follows: the length, width and thickness of the metallic nanofilm A are $L_1=12\mu m$, $a_1=800nm$ and $d_1=100nm$. The length, width and thickness of graphene are $L_2=4\mu m$, $a_2=3\mu m$ and $d_2=0.34nm$, and the parameter of the metallic nanofilm B is the same as A.

3.1. Numerical simulation of electrical conductivity and uncertainty analysis.

Fig. 7 The electric field of the whole H-type device at $U_2 = 0.05V$.

A numerical simulation was carried out by using COMSOL Multiphysics Software to calculate the electric field of the whole H-type device as shown in Fig.7, where the electrical conductivity of graphene was set to $\sigma = 4\times10^5\Omega^{-1}m^{-1}$ and the voltage U_2 across graphene was 0.05 V.

According to the Equation (1) of calculating electrical conductivity, it is known that the sources of uncertainty

mainly include the uncertainty of geometric dimensions of the 2D material, and the uncertainty of voltage and current measurement. The geometric dimensions of the 2D material were measured by SEM imaging and the relative error is about 0.15%. The voltage and current were measured by a high-precision digital multimeter (Keithley 2002) with a relative error of 0.01%. Therefore, the total uncertainty should be less than 0.2%. The contact resistance between the graphene and metallic nanofilms is also an important issue. We give some discussion of contact resistance in the [supplementary material](#).

3.2 Numerical simulation of thermal conductivity and uncertainty analysis

Fig. 8 The temperature distribution of the whole H-type device at given properties from literature.

Fig. 8 shows the simulation of thermal conductivity, where the thermal conductivity of graphene was set to $\lambda_2=2400 \text{ Wm}^{-1}\text{K}^{-1}$ and the voltage U_1 across metallic nanofilm A was 0.06 V. The main sources of uncertainty in the thermal conductivity measurement include the uncertainty of geometric dimensions of 2D material, the uncertainty of temperature measurement through nanofilm as resistance thermometer, the uncertainty of numerical calculation, the uncertainty of contact thermal resistance R_C between nanofilm and 2D material and the effects of thermal radiation and convection. The total uncertainty of thermal conductivity measurement is about 7.5%. See the [supplementary material](#) for more details about the uncertainty analysis of each part.

3.3 Numerical simulation of Seebeck coefficient and uncertainty analysis

Assume that the laser absorption rate η is 0.02. Laser power $q=5 \text{ mW}$. Laser radius $r=0.5 \mu\text{m}$. The Seebeck coefficient is set to be $70 \mu\text{V/K}$ as recommended in the literature. A relatively small voltage was applied to both metallic nanofilms and the Joule heating effect could be neglected. In the simulation, several positions were set for laser heating as

$c = 0.5, 0.6, 0.7, 0.8 \mu\text{m}$, where c is the distance between the middle point of nanofilm A and the center of laser spot. **Fig. 9** shows the temperature distribution of H-type device at $c = 0.6 \mu\text{m}$, and the temperature difference of graphene is 5.66 K .

Fig. 9 The temperature field distribution of H-type device heated by laser beam at $c = 0.6 \mu\text{m}$, $q = 5 \text{ mW}$ and $\eta = 0.02$.

The sources of uncertainty in the Seebeck coefficient measurement mainly include the uncertainty of positioning the laser spot, numerical calculations, potential difference measurement, temperature measurement through nanofilm as resistance thermometer, the contact thermal resistance R_C between nanofilm and 2D material, the thermal conductivity of 2D material and the effects of thermal radiation and convection. The total uncertainty of Seebeck coefficient measurement is about 12%. The Seebeck coefficient is sensitive to the temperature difference across the 2D material, but not to the absolute temperature. Therefore, the uncertainty of Seebeck coefficient is smaller than that of thermal conductivity. Besides, how to control the laser spot size as well as the laser intensity distribution is also an important question. See the [supplementary material](#) for more details about the uncertainty analysis of each part.

4. Conclusions

This work proposes a new comprehensive method to measure the thermoelectric properties of 2D materials. The electrical conductivity, thermal conductivity and Seebeck coefficient of free-standing 2D material can be measured in-situ on the same sample by simply changing the external electrical circuit. This method can provide full thermoelectric properties of a pristine 2D material with high precision and the experimental procedures is easy to perform. Meanwhile, the laser absorption rate of 2D material can be obtained as well. Graphene was used as an example to demonstrate the feasibility of current method. The recommended properties of graphene in literature were used to perform uncertainty analysis. This method and measurement results are useful to promote the development of new 2D thermoelectric devices.

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Conflict of interest

There are no conflicts to declare.

Supporting information

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