Supplementary material

An integrated H-type method to Measure thermoelectric properties of
two-dimensional materials.

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1. Mathematical analysis in the thermal conductivity measurement

![Diagram](image)

**Fig. 1.** 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.

Fig. 1 shows the 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 as follows:

\[
\begin{align*}
\text{metallic nanofilm A:} & \quad \frac{d^2 T_1(x)}{dx^2} + \frac{U_1 I_1}{L_1 a_1 d_1 \lambda_1} - \frac{T_1(x) - T_2(x,0)}{d_2 \lambda_2 R_C} = 0, \quad 0,0 \leq x \leq \frac{L_2}{2} \\
& \quad \frac{d^2 T_1(x)}{dx^2} + \frac{U_1 I_1}{L_1 a_1 d_1 \lambda_1} = 0, \quad \frac{L_2}{2} < x \leq \frac{L_1}{2} \\
\text{2D material:} & \quad \frac{\partial^2 T_2(x,y)}{\partial x^2} + \frac{\partial^2 T_2(x,y)}{\partial y^2} = 0 \\
& \quad \frac{d^2 T_3(x)}{dx^2} - \frac{T_2(x) - T_3(x,a_2)}{d_3 \lambda_3 R_C} = 0, \quad 0,0 \leq x \leq \frac{L_3}{2} \\
& \quad \frac{d^2 T_3(x)}{dx^2} = 0, \quad \frac{L_2}{2} < x \leq \frac{L_3}{2}
\end{align*}
\]

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. \(\lambda_1\) and \(\lambda_3\) 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.\(^{[1,2]}\)

It is impossible to directly calculate the thermal conductivity. Through the numerical simulation of the Eq. (1), we found that the temperature distribution of metallic nanofilm A can be approximated as follow:
\( T_i(x) = \begin{cases} Jx^4 + Kx^2 + M, & 0 \leq x < \frac{L_1}{2} \\ -\frac{s}{2} x^2 + Nx + \frac{L_2}{2}, & \frac{L_1}{2} \leq x \leq \frac{L_1}{2} \end{cases} \) (2)

where, \( J, K, N, W \) are the unknown parameters and \( s = \frac{U_1 h_1}{(L_1 a_1 d_1 \lambda_1)} \).

The boundary condition for \( T_i(x) \) at \( x = 0 \) is:

\[
\left. \frac{dT_i(x)}{dx} \right|_{x=0} = (3)
\]

\( T_i(x) \) is continuous and derivable at \( x=L_2/2 \):

\[
J \left( \frac{L_2}{2} \right)^4 + K \left( \frac{L_2}{2} \right)^2 + M = -\frac{s}{2} \left( \frac{L_2}{2} \right)^2 + N \frac{L_2}{2} + W \quad (4)
\]

\[
4J \left( \frac{L_2}{2} \right)^3 + 2K \frac{L_2}{2} = -2s \frac{L_2}{2} + N \quad (5)
\]

The boundary conditions at \( x=L_1/2 \) is:

\[
T_i \left( \frac{L_1}{2} \right) = -\frac{s}{2} \left( \frac{L_1}{2} \right)^3 + N \frac{L_1}{2} + W = T_o \quad (6)
\]

In the thermal conductivity measurement, we can know that the average temperature of metallic nanofilm A is:

\[
\frac{1}{L_1/2} \int_0^{L_1/2} T_i(x)dx = T_{a1} \quad (7)
\]

According to Eq. (1) and Eq. (2), the \( T_2(x,0) \) can be expressed as:

\[
T_2(x,0) = Jx^4 + Kx^2 + M - (12Jx^2 + K + s)R_c d_i \lambda_i \quad (8)
\]

The boundary condition for \( T_2(x, y) \) at \( x = 0 \) is:

\[
\left. \frac{\partial T_2(x, y)}{\partial x} \right|_{x=0} = 0 \quad (9)
\]
The boundary condition for $T_2(x, y)$ at $x = L_2/2$ is:

$$\frac{\partial T_2(x, y)}{\partial x} \bigg|_{x=L_2/2} = 0$$  \hspace{1cm} (10)

The boundary condition for $T_2(x, y)$ at $y = 0$ are:

$$T_2(x, 0) = Jx^4 + Kx^2 + M - (12Jx^2 + K + s)R_c \frac{d}{d} \lambda_1$$  \hspace{1cm} (8)

$$\frac{\partial T_2}{\partial y} \bigg|_{y=0} = a_1 \frac{T_2(x, 0)}{-\lambda_2 d R_c} = a_1 \frac{(12Jx^2 + K + s) d}{-\lambda_2 d_2}$$  \hspace{1cm} (11)

Then $T_2(x, y)$ can be obtained as:

$$T_2(x, y) = \sum_{n=1}^{\infty} \left[ C_n \exp(-\frac{n\pi}{L_2/2} y) + D_n \exp(-\frac{n\pi}{L_2/2} y) \right] \cos(\frac{n\pi}{L_2/2} x)$$  \hspace{1cm} (12)

Where $C_n$ and $D_n$ are:

$$C_n = \frac{1}{L_2/2} \left[ \int_0^{L_2/2} T_2(t, 0) \cos(\frac{n\pi}{L_2/2} t) dt + \frac{L_2/2}{n\pi} \int_0^{L_2/2} a_1 \frac{T_2(t) - T_2(t, 0)}{-\lambda_2 d R_c} \cos(\frac{n\pi}{L_2/2} t) dt \right]$$  \hspace{1cm} (13)

$$D_n = \frac{1}{L_2/2} \left[ \int_0^{L_2/2} T_2(t, 0) \cos(\frac{n\pi}{L_2/2} t) dt - \frac{L_2/2}{n\pi} \int_0^{L_2/2} a_1 \frac{T_2(t) - T_2(t, 0)}{-\lambda_2 d R_c} \cos(\frac{n\pi}{L_2/2} t) dt \right]$$  \hspace{1cm} (14)

And then we can know $T_2(x, a)$ as:

$$T_2(x, a_2) = \sum_{n=1}^{\infty} \left[ C_n \exp(-\frac{n\pi}{L_2/2} a_2) + D_n \exp(-\frac{n\pi}{L_2/2} a_2) \right] \cos(\frac{n\pi}{L_2/2} x)$$  \hspace{1cm} (15)

Substitute Eq. (15) into Eq. (1) to get $T_3(x)$:

$$T_3(x) = \begin{cases} E \exp\left(\frac{1}{d_3 \lambda_3 R_c} x\right) + F \exp\left(-\frac{1}{d_3 \lambda_3 R_c} x\right) + \phi(x), & 0 \leq x \leq \frac{L_2}{2} \\ Gx + H, & \frac{L_2}{2} < x \leq \frac{L_2}{2} \end{cases}$$  \hspace{1cm} (16)

Where, $E$, $F$, $G$, $H$ are the unknown parameters.
\[ \phi(x) = \sum_{n=1}^{\infty} \frac{1}{d_n \lambda_n R_c} \left[ C_n \exp\left( \frac{-n\pi}{L_x} a_x \right) + D_n \exp\left( \frac{-n\pi}{L_x} a_x \right) \right] \cos\left( \frac{n\pi}{L_x} x \right)/\left[ \frac{1}{d_n \lambda_n R_c} + \left( \frac{n\pi}{L_x} \right)^2 \right] \] (17)

The boundary condition for \( T_3(x) \) at \( x = 0 \) is:

\[ \frac{dT_3(x)}{dx} \bigg|_{x=0} = 0 \] (18)

\( T_3(x) \) is continuous and derivable at \( x=L_2/2 \):

\[ E \exp\left( \frac{1}{d_3 \lambda_3 R_c} \frac{L_2}{2} \right) + F \exp\left( -\frac{1}{d_3 \lambda_3 R_c} \frac{L_2}{2} \right) + \phi\left( \frac{L_2}{2} \right) = G \frac{L_2}{2} + H \] (19)

\[ \sqrt{\frac{1}{d_3 \lambda_3 R_c}} \left[ E \exp\left( \frac{1}{d_3 \lambda_3 R_c} \frac{L_2}{2} \right) - F \exp\left( -\frac{1}{d_3 \lambda_3 R_c} \frac{L_2}{2} \right) \right] + \frac{d\phi}{dx} \bigg|_{x=L_2/2} = G \] (20)

The boundary conditions for \( T_3(x) \) at \( x=L_3/2 \) is:

\[ G \frac{L_3}{2} + H = T_0 \] (21)

Based on energy conservation, we can know that:

\[ -\lambda_2 a_2 d\chi \frac{\partial T_2}{\partial y} \bigg|_{y=a_2} = a_2 d\chi \frac{T_2(x, a_2) - T_2(x)}{R_c} \] (22)

We can also know that the average temperature of metallic nanofilm B is:

\[ \frac{1}{L_3 / 2} \int_{0}^{L_3 / 2} T_3(x) dx = T_{\text{avg}} \] (23)

There are ten unknow parameters: \( J, K, M, N, W, E, F, G, H \) and \( \lambda_2 \), and there are ten equations as well: Eq. (3-6) and Eq. (18-23). We can solve all unknown parameters and obtain the temperature distribution of \( T_1(x) \), \( T_2(x, y) \) and \( T_3(x) \) through MATLAB.
The thermal conductivity of 2D material can be expressed as:

$$\lambda_2 = -\frac{T_1(x) - T_2(x, 0)}{R_c \frac{\partial T_2}{\partial y}_{y=0}}$$

(24)

2. The uncertainty analysis of thermal conductivity

The main sources of uncertainty in the thermal conductivity measurement include the uncertainty of the numerical calculation accuracy, geometric dimensions of 2D material, the effects of thermal radiation and convection, the temperature measurement through nanofilm as resistance thermometer, and the contact thermal resistance $R_c$ between nanofilm and 2D material.
Fig. 2 shows the situation of grid encryption in numerical simulation. After grid encryption, we found that both the temperature errors of metallic nanofilms A and B are less than 0.001K.

The geometric dimensions of the sample were measured by SEM imaging and the relative error is about 0.1%. By changing the geometric dimensions, we found that both the temperature errors of metallic nanofilms A and B are less than 0.001K.

The experiment was carried out in a high vacuum chamber ($10^{-4}$Pa). Therefore, the impact of convection is negligible. By introducing radiation in the numerical simulation,
there are about 0.0003K temperature error.

The well calibrated nanofilm sensor can offer a temperature measurement accuracy better than 0.01K.

The contact thermal resistance $R_C$ between nanofilm and 2D material may cause important effect to numerical simulation. Table 1 shows the effect of $R_C$ on the temperature of metallic nanofilms A and B. According to the literature, the contact thermal resistance $R_C$ between Au nanofilm and graphene is about $10^{-8}$ Km²W⁻¹, resulting in less than 0.08K temperature error.

<table>
<thead>
<tr>
<th>$R_C$/Km²W⁻¹</th>
<th>$T_{3A}$/K</th>
<th>$T_{3B}$/K</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0E-07</td>
<td>313.302</td>
<td>294.531</td>
</tr>
<tr>
<td>1.0E-08</td>
<td>313.154</td>
<td>294.671</td>
</tr>
<tr>
<td>1.0E-09</td>
<td>313.102</td>
<td>294.720</td>
</tr>
<tr>
<td>1.0E-10</td>
<td>313.084</td>
<td>294.736</td>
</tr>
<tr>
<td>1.0E-11</td>
<td>313.080</td>
<td>294.740</td>
</tr>
<tr>
<td>1.0E-12</td>
<td>313.080</td>
<td>294.741</td>
</tr>
<tr>
<td>0</td>
<td>313.080</td>
<td>294.741</td>
</tr>
</tbody>
</table>
The sum of all the above errors is less than 0.09K. Fig. 3 and Fig. 4 show the effect of thermal conductivity $\lambda_2$ on the average temperature of metallic nanofilms A and B. If $T_{\lambda A}$ or $T_{\lambda B}$ change 0.01K, the $\lambda_2$ will change $20\text{Wm}^{-1}\text{K}^{-1}$. In our measurement the total uncertainty of $\lambda_2$ is $0.09/0.01\times20/2400=7.5\%$

Fig. 3. The effect of $\lambda_2$ on $T_{\lambda A}$. 

$T_{\lambda A} = -0.0006\lambda_2 + 314.42$

$R^2 = 0.9999$

$T_{\lambda A} = 313.08\text{K}$
3. **The uncertainty analysis of Seebeck coefficient**

The sources of uncertainty in the Seebeck coefficient measurement mainly include the uncertainty of positioning the laser spot, numerical calculation accuracy, the effects of thermal radiation and convection, potential difference measurement, temperature measurement through nanofilm as resistance thermometer, the contact thermal resistance $R_C$ between nanofilm and 2D material and the thermal conductivity of 2D material.

The uncertainty from positioning the laser spot is not negligible. The accuracy of positioning the laser spot is 50nm. By comparing the

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**Figure 4.** The effect of $\lambda_2$ on $T_{AB}$. 

$$T_{AB} = 0.0005\lambda_2 + 293.43$$

$$R^2 = 0.9999$$
temperature difference \((T_{2A} - T_{2B})\) before and after changing the laser spot position in the simulation, we got the effect on temperature difference, which is about 6\%-7%.

The source of uncertainty from numerical calculations accuracy and the effects of thermal radiation and convection can be ignored like the uncertainty of thermal conductivity. The uncertainty of potential difference measure is about 0.02%, which is ignorable.

Table 2. shows the effect of \(R_C\) on the temperature difference \((T_{2A} - T_{2B})\). The uncertainty caused by \(R_C\) is about 1.6% when \(R_C = 10^{-8}\text{Km}^2\text{W}^{-1}\). Fig. 5 shows the effect of the thermal conductivity of 2D material on \((T_{2A} - T_{2B})\), which cause an uncertainty less than 1.7% when thermal conductivity changes 10%. The temperature rise of \(T_{2A}\) is about 0.6 times of the temperature rise of the \(T_{5A}\). The well calibrated nanofilm sensor can offer a temperature measurement accuracy better than 0.01K. Therefore, the accuracy of the \(T_{2A}\) is 0.006K, resulting in an uncertainty of 0.93%. \(T_{2B}\) is the same as \(T_{2A}\), having an uncertainty of 0.93%. The total uncertainty is about 7%+1.6%+1.7%+0.93%+0.93% \approx 12\%.
Table 2. The effect of $R_C$ on $(T_{2A} - T_{2B})$ and uncertainty from $R_C$

<table>
<thead>
<tr>
<th>$R_C$/Km$^2$W$^{-1}$</th>
<th>$(T_{2A} - T_{2B})$/K</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0E+07</td>
<td>6.469</td>
<td>14.2%</td>
</tr>
<tr>
<td>1.0E+08</td>
<td>5.572</td>
<td>1.6%</td>
</tr>
<tr>
<td>1.0E+09</td>
<td>5.598</td>
<td>1.1%</td>
</tr>
<tr>
<td>1.0E+10</td>
<td>5.645</td>
<td>0.3%</td>
</tr>
<tr>
<td>1.0E+11</td>
<td>5.658</td>
<td>0.0%</td>
</tr>
<tr>
<td>1.0E+12</td>
<td>5.660</td>
<td>0.0%</td>
</tr>
<tr>
<td>0</td>
<td>5.660</td>
<td>0.0%</td>
</tr>
</tbody>
</table>

Fig. 5. The effect of thermal conductivity of 2D material on $(T_{2A} - T_{2B})$.

4. The effect of metallic voltage drops on Seebeck coefficient.
The signal V between two electrodes in measuring Seebeck coefficient includes the metallic voltage drop and the voltage of the 2D material. The resistance of the 2D material such as graphene is about 5500 ohms, while the resistance of the metallic nanofilm in the entire circuit is about 2.4 ohms, which can be ignored.

5. Preparation of suspended H-type graphene device

As shown in Fig.6, a free-standing graphene can be prepared following a series of Micro-Electro-Mechanical System (MEMS) process discussed in our previous paper[3,4,5]

Fig. 6. SEM images of the fabricated free-standing H-type graphene. This figure is reprinted with permission from Springer Nature publishing group.
6. Discussion on the effect of contact resistance on electrical conductivity

The contact resistance between the graphene and metallic nanofilms may have effects on electrical conductivity. However, it is difficult to measure the contact resistance accurately, which may require the transfer length method (TLM).\cite{6,7} Also, the value of contact resistance highly depends on the condition of metal/graphene interface. That is why the reported data for contact resistance are scattered. As far as we know, in some published papers on the electrical properties of graphene, the contact resistance has not been precisely measured and considered in the analysis due to this difficulty.\cite{8-10} The paper aims to propose a feasible experimental method for comprehensive characterization of three properties ($\sigma$, $\lambda$, $S$) of the same 2D material sample. The measurement of electrical contact resistance is not the main purpose of this work. Also, it needs to be mentioned that the metallic nanofilms were directly deposited on the 2D material, which can significantly enhance the binding force, avoid the influence of impurities and defects and further reduce the contact resistance.

7. How to control the laser spot size as well as the laser intensity distribution?

J. H. Liu et al proposed a method that can measure the laser spot size and laser intensity distribution.\cite{11} The laser spot size and intensity distribution was determined by fitting the Raman signal profile obtained by scanning across an individual SWCNT with
very small step size, as shown in Fig. 7. An example was shown in Fig. 8.

We will calibrate the laser spot size and laser intensity distribution before the experiment by this method, thus the uncertainty from the laser spot and laser intensity distribution can be ignored.

**Fig. 7.** Focused laser spot scanning across an individual suspended SWCNT. This figure is reprinted with permission from Royal Society of Chemistry.
Fig. 8. Raman signal intensity profile. The focused laser spot was scanned across the SWCNT along the direction perpendicular to the SWCNT axis with 0.1 μm step size to measure the Raman intensity profile shown as the squares in Fig. 8. The Gaussian peak function $I=405\exp\left(-\frac{x}{0.53}^2\right)$ and $I=405\exp\left(-\frac{x}{0.45}^2\right)$ fit well with the experimental results. Thus, the experimental results indicate the laser spot profile is $I=405\exp\left(-\frac{x}{0.49}^2\right)$. This figure is reprinted with permission from Royal Society of Chemistry.

References