Thermoelectric Parameter Modeling of Single-Layer Graphene Considering Carrier Concentration and Mobility With Temperature and Gate Voltage

NING WANG, CONG MENG, ZHI-HAO MA, CONG GAO, HONG-ZHI JIA, GUO-RONG SUI, AND XIU-MIN GAO
Shanghai Key Laboratory of Modern System, Engineering Research Center of Optical Instrument and System, Ministry of Education, University of Shanghai for Science and Technology, Shanghai 200093, China

Corresponding author: Xiu-Min Gao (xiumin_gao@aliyun.com)

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Abstract

Single-layer graphene (SLG) sheets can exhibit thermoelectric properties under the control of gate voltage. The controlled factors and regulation mechanism of SLG thermoelectric properties have become research hotspots. In this paper, a SLG thermoelectric parameter model considering carrier concentration and mobility with temperature and gate voltage is proposed. Based on the proposed model, the square resistance ($R_s$) and Seebeck coefficient ($S$) of the SLG are calculated. The results show that the maximum value of $R_s$ decreases from 5.8 K$\Omega$ to 3.2 K$\Omega$ at the Dirac voltage when the temperature increases from 100 K to 500 K. A large and stable $S$ can be obtained at high voltages and temperatures. The maximum value of $S$ can reach 161.3 $\mu$V/K at $T$ = 500 K, exhibiting a more obvious thermoelectric characteristic. Simultaneously, the saturation law of the power factor ($Q$) with the change of gate voltage and the amplitude regulation of $Q$ by temperature are obtained. This work can provide a theoretical basis for analyzing the thermoelectric characteristics of SLG.

Index Terms

Graphene, seebeck coefficient, square resistance, thermoelectric effect.

I. INTRODUCTION

The thermoelectric effect is the direct conversion of heat into electricity or electricity into heat and includes the Seebeck effect, the Peltier effect and the Thomson effect [1]. As a carrier of the thermoelectric effect, the thermoelectric material realizes the mutual transformation between electricity and heat [2]. Recently, with the emergence of new thermoelectric materials [3], low-dimensional thermoelectric materials exhibit excellent electrical and thermal characteristics and have attracted attention from researchers. Graphene is one of these new low-dimensional thermoelectric materials.

Graphene, a new two-dimensional thin film material composed of carbon atoms, has a thickness of only one carbon atom (0.34 nm). Since it was first discovered in 2004 [4], graphene has become the most attractive nanomaterial due to its excellent electrical, thermal and optical properties [5]–[9]. Initially, research on graphene materials was not carried out in the field of thermoelectric but focused on light, heat and electricity. For example, a research team from Nankai University confirmed that graphene exhibits strong polarization-dependent optical absorption under total internal reflection [10], that is, graphene can be used as a sensitive material for fiber-optic sensors, which makes it possible to replace the precious metal layer in fiber sensors with a graphene layer [11]. In addition, the thermal conductivity of SLG has been measured up to 5300 W/mK by using Raman spectroscopy [12]. This property makes graphene an excellent material for thermal management and provides additional power for the integration of CMOS devices and circuits. In the temperature environment of 5 K, the electron mobility in SLG is captured up to 200,000 cm$^2$/Vs [13]. Hence, it can be used as a high-speed conduction channel in signal transmission. In addition to the above fields, the research and application
of graphene as a thermoelectric material are being explored gradually. However, early research on the ability of graphene to play a role in thermoelectric energy conversion is not optimistic. Experiments by Michael S. Fuhrer et al. show that temperature has a limited effect on electron mobility in graphene [14]. Its Seebeck coefficient is very low due to the zero band gap of ideal graphene [15], which makes graphene unsuitable as a thermoelectric material. It was not until 2007 that this streak was broken by Melinda Y. Han. Han found that the electron band gap of graphene is inversely proportional to the width of graphene sheets [16]. That is, the gap of graphene has physical regulation characteristics. Therefore, graphene nanoribbons with open electron gaps can be obtained by changing the width of the graphene [17], [18]. Experiments have confirmed that the carrier mobility of graphene changes with the generation of its band gap [19]. However, this scale regulation is difficult to use flexibly after fabrication. K. S. Novoselov’s team found that graphene exhibited a bipolar electrical tuning effect when an external bias voltage was applied. A large number of electrons (holes) are induced by the positive (negative) gate voltage, and all of the induced carriers are movable [20], which lays the foundation for regulating the graphene semiconductor characteristics through voltage. Conversely, the carrier mobility of single-layer, double-layer and triple-layer graphene and their temperature dependence were systematically studied by Zhu’s team [21]. The mobility of SLG increases with the increase of temperature and carrier density. Using silicon dioxide (SiO$_2$) as the substrate, Dorgan et al. [22] obtained changes in the SLG carrier mobility and saturation velocity with temperature. Ouyang et al. [23] solved the atomic electron and phonon transport in the form of the non-equilibrium Green’s function, which caused the Seebeck coefficient to be large. At a temperature of 300 K, $S$ is calculated to be a maximum of 90 $\mu$V/K [24]. It can be seen that in addition to voltage, temperature can be used as an additional control means to regulate the carrier and thermoelectric parameters of graphene.

The studies mentioned above provide a theoretical basis for exploring the electrical and thermoelectric properties of graphene. The carrier concentration and mobility of SLG, including the thermoelectric parameters, are regulated by the temperature and gate voltage. In this paper, a model of the SLG thermoelectric parameters that considers the carrier concentration and mobility at various temperatures and gate voltages is proposed. The regulation of $R_s$ by different gate voltages and temperatures is discussed. The thermoelectric parameters $S$, $Q$ and $V_s$ of SLG are calculated, and the adjustment law of the gate voltages and temperatures on these parameters is explored.

II. ELECTRICAL CHARACTERISTICS FOR SLG

Fig. 1 shows the device structure of SLG in its operating state. In this structure, SiO$_2$ is used as the intermediate oxide layer, and a highly doped silicon substrate is used as the back gate. The carrier concentration in the SLG can be regulated by the vertical electric field applied by the back gate voltage ($V_G$). $V_s$ is the Seebeck voltage. A potential difference will be produced between two dissimilar electrical conductors or semiconductors if there is a temperature gradient across them [25], and this voltage difference is called the Seebeck voltage. $l_g = 7 \mu$m, $w_g = 4 \mu$m are the length and width of the graphene nanoribbon, respectively. In this section, the variations of the carrier concentration and the mobility in the SLG are studied, and the square resistance of the SLG is calculated to investigate the electrical properties of SLG under an electric field.

A. CARRIER CONCENTRATION

The relationship between the gate voltage and the carrier concentration ($n_g$) induced by the gate voltage can be obtained from [21]

$$n_g = (V_G - V_0) / \left( q \left( \frac{1}{C_{ox}} + \frac{1}{C_q} \right) \right) \quad (1)$$

where $C_{ox} = \varepsilon_0 \varepsilon_r / t_{ox}$ is the oxide capacitance, $\varepsilon_0$, $\varepsilon_r$ and $t_{ox}$ are the vacuum dielectric constant, the relative dielectric constant and the thickness of SiO$_2$, respectively. $C_q$ is the quantum capacitance of graphene, and $q$ is the electron charge. For SiO$_2$ with a thickness of 300 nm, $C_q$ in (1)
At the Dirac point of SLG is in the valence band. As that, the Fermi level is at the Dirac point, whereas short-range scattering is proportional to the carrier concentration.

\[ n = \sqrt{\left(\frac{C_{ox} V_{G0}}{q}\right)^2 + 4 \left(\frac{n^*}{2}\right)^2 + n_{th}^2} \]  

where \( n^* \) is the residual carrier puddle density, and \( n_{th} \) is the thermal carrier density. \( n_{th} = \frac{\pi}{6}(\hbar v_F)^2 \), where \( v_F \) and \( \hbar \) are the Fermi velocity and the reduced Planck constant, respectively. \( n \) contains \( n_{th} \) and \( n_g \). The line in Fig. 2 indicates that \( T \) has a positive effect on \( n_{th} \). In Fig. 3, the dependence of \( n \) on \( T \) and \( V_{G0} \) is depicted. When \( V_{G0} \approx 0 \) V, the Fermi level of SLG is in the valence band. As \( V_{G0} \) approaches 0 V, the Fermi level moves from the valence band to the Dirac point. At the Dirac point (\( V_{G0} = 0 \) V), the carrier concentration in the SLG channel is the lowest. When \( V_{G0} > 0 \) V, the Fermi level enters the conduction band, and the carrier concentration in the SLG channel also begins to rise. At the same reference temperature, the larger the absolute value of \( V_{G0} \), the higher the \( n \). The Gate-only curve denotes the relationship between \( n_g \) and \( V_{G0} \). \( n_g \) is close to zero at \( V_{G0} \approx 0 \) V, but \( T \) has a positive effect on \( n_{th} \). In the case of \( V_{G0} = 2 \) V where \( n_g = 0.15 \times 10^{12}\text{cm}^{-2} \), \( n = 0.16 \times 10^{12}\text{cm}^{-2} \) at \( T = 100 \) K, whereas \( n \) can reach \( 0.52 \times 10^{12}\text{cm}^{-2} \) at \( T = 500 \) K. \( n \) has risen more than three times under the regulation of \( T \). However, \( n_g = 1.5 \times 10^{12}\text{cm}^{-2} \) when \( V_{G0} = 20 \) V. Meanwhile, \( n = 1.51 \times 10^{12}\text{cm}^{-2} \) at \( T = 100 \) K, and \( n = 1.59 \times 10^{12}\text{cm}^{-2} \) at \( T = 500 \) K. Thus, in the low gate voltage range, \( n_{th} \) plays a leading role on \( n \). Whereas the carrier concentration curves at different temperatures gradually coincide with the Gate-only curve with the increase of \( V_{G0} \), indicating that \( n_g \) begins to dominate. This suggests that \( n \) is subject to different main control mechanisms under different conditions, and this result is consistent with reference [22]. The desired carrier concentration can be achieved by equalizing \( T \) and \( V_{G0} \).

**B. CARRIER MOBILITY**

The carrier mobility (\( \mu \)) refers to the overall speed of electrons and holes in the semiconductor. To simplify the calculation, both \( \mu \) and \( n_0 \) have been used as constant values for subsequent calculations in many studies [26], [27]. In fact, \( \mu \) will also change as the conditions imposed by the system change [28].

\[
\mu = \frac{\mu_0}{1 + \left(\frac{\sqrt{(\frac{C_{ox} V_{G0}}{q})^2 + 4 (\frac{n^*}{2})^2 + n_{th}^2}}{n_{ref}}\right)^\alpha} \times \frac{1}{1 + (\frac{T}{T_{ref}} - 1)^\beta}
\]

where \( \mu_0 \) is the low field mobility, \( T_{ref} \) and \( n_{ref} \) are the reference temperature and the reference charge density [29], respectively, and \( \alpha \) and \( \beta \) are constants with values of 2.2 and 3, respectively. The values of \( T_{ref}, n_{ref}, \alpha \) and \( \beta \) are derived from the traditional model, and we take them from [22], [29]. In (3), \( \mu \) is negatively correlated with \( T \) and \( V_{G0} \).

In Fig. 4, the value of \( \mu \) is plotted as \( V_{G0} \) and \( T \) vary. The peak value of \( \mu \) is 6610 cm²/Vs at \( T = 100 \) K, which is smaller than the carrier mobility in pure graphene sheets. Since the scattering effect of the carrier by the optical phonon in the SiO₂ film is much larger than the one by the SLG itself, a large drop in \( \mu \) occurs, and \( n \) of the SLG is enhanced.

A high \( T \) and a large \( V_{G0} \) lead to a decrease in \( \mu \), which is consistent with [22]. Based on the work in [21], when the temperature is low, such as \( T = 100 \) K, \( \mu \) is mainly limited by Coulomb scattering and short-range scattering. Coulomb scattering is independent of the carrier concentration, whereas short-range scattering is proportional to the carrier concentration. Therefore, for the curve of \( T = 100 \) K, Coulomb scattering is enhanced when \( V_{G0} \) is adjusted to increase the carrier concentration, and \( \mu \) decreases. \( \mu \) drops
Combined with (2), the expression of et al. proposed by Grosse and the above results, both $n$ simplifies the carrier concentration and mobility. Based on temperatures, $\mu$ surface polar phonons on the SiO$_2$ scattering of SLG is proportional to $T$. The formula for calculating the square resistance, $\mu$ however, $\mu$ drops sharply by $\Delta \mu_3 = 898$ cm$^2$/Vs as $T$ continues to rise from 400K to 500K. Since the acoustic phonon scattering of SLG is proportional to $T$ and the scattering of surface polar phonons on the SiO$_2$ substrate is excited by high temperatures, $\mu$ is further attenuated.

C. SQUARE RESISTANCE

The formula for calculating the square resistance, $R_s = 1/q\mu n$, was established according to the model proposed by Grosse et al. [30]. However, this expression simplifies the carrier concentration and mobility. Based on the above results, both $n$ and $\mu$ are affected by $T$ and $V_{GO}$. Combined with (2), the expression of $R_s$ can be written as

$$R_s = \frac{(1 + (n/n_{ref})^{\alpha})(1 + (T/T_{ref} - 1)^{\beta})}{-q\mu_0 n}$$

(4)

To introduce the two variables $T$ and $V_{GO}$, $n_{th}$ is used in (3), and $R_s$ can be obtained by combining (3) and (4):

$$R_s = \frac{1 + \left(\frac{C_0 (V_{GO})}{q}\right)^2 + 4\left(\frac{n}{T}\right)^2 + \left(\frac{\pi}{6} \left(\frac{\mu_0 T}{\nu} \right)^2\right)^2/n_{ref}}{-q\mu_0}$$

$$\times \left(1 + (T/T_{ref} - 1)^{\beta}\right)^{\alpha}$$

(5)

In (5), once the material used is given, many parameters in the formula are also determined. As a result, $R_s$ is only controlled by the two variables $T$ and $V_{GO}$. Fig. 5 depicts the relationship of $R_s$ and $V_{GO}$ at different temperatures. $R_s$ is greatly affected by $T$ at low $V_{GO}$ due to the dominant position of $n_{th}$. Moreover, the value of $R_s$ at different temperatures reaches the maximum at $V_{GO} = 0$ V. $n$ is the smallest at this point, and $R_s$ is inversely proportional to $n$. Note that the peak value of $R_s$ can reach 5.3 KΩ at $T = 100$ K, whereas the peak value is only 3.3 KΩ at $T = 500$ K. From these results, the effect of $T$ on $R_s$ cannot be ignored when $V_{GO}$ is in a low range. However, the dominant position is occupied by $n_{th}$ as $V_{GO}$ rises gradually, which means that $R_s$ begins to be regulated by $V_{GO}$. With the increase of $T$, the lattice vibration is strengthened, and the phonon motion is intensified, resulting in an increase in scattering. Therefore, in the case of a large $V_{GO}$, a higher $T$ corresponds to a larger $R_s$.

Since conductance is inversely proportional to $R_s$, Fig. 6 can be obtained by combining with (4). Fig. 6 shows the relationship of $R_s$ and conductance as a function of $n$ at different temperature conditions. $R_s$ rises sharply when $n$ is less than $0.5 \times 10^{12}$cm$^{-2}$, and therefore, SLG is not suitable for use as a thermoelectric device if the internal
carrier concentration is too small. As $n$ becomes larger, the effect of $T$ on $R_c$ gradually weakens, whereas the impact on conductance begins to be apparent. The conductance and $n$ are approximately linearly related, contrary to the relationship between $R_c$ and $n$. Similarly, high $T$ values exacerbate the scattering of acoustic phonons and surface polar phonons on the SiO$_2$ substrate, reducing $\mu$. The conductance is proportional to $\mu$, and thus, the conductance is reduced as $T$ increases. The conductance can be altered by adjusting the internal carrier concentration of SLG, and the carrier concentration can be regulated by modifying the gate voltage, which provides a more convenient form for the conductivity revision of SLG. The purpose of increasing the conductance of SLG can also be achieved by adjusting the $T$ of $\mu$ and $n$.

### III. HERMOELECTRIC CHARACTERISTICS OF SLG

The Seebeck effect, also known as the first thermoelectric effect, is a thermoelectric phenomenon in which $V_S$ is produced. However, the performance of a thermoelectric material is not usually determined by $V_S$; rather, it is determined by the thermoelectric figure of merit $ZT$ [31].

$$ZT = \frac{S^2 \sigma T}{K}$$  

(6)

where $S$ and $K$ are the Seebeck coefficient and the thermal conductivity, respectively. $\sigma = l_g/(R_c w d)$ is the conductivity, and $d$ is the thickness of the SLG. Generally, the larger the $ZT$ value, the better the thermoelectric properties of the material. It can be seen from (6) that it is necessary to increase $S$ and $\sigma$ or to weaken $K$ to improve the performance of SLG.

#### A. SEEBECK COEFFICIENT

According to the Mott relationship [32]–[34], the expression of $S$ can be obtained as follows:

$$S = -\frac{2\pi^2}{3} \frac{K_B^2 T}{q} \frac{dG}{dE} = \frac{\pi^2}{3} \frac{K_B^2 T}{q} \frac{dR}{dE}$$  

(7)

where $K_B$ and $E = E_F$ are the Boltzmann constant and the Fermi level, respectively. The dependence of $E_F$ on the carrier density can be acquired from the tight binding calculation [35]. For SLG, $E_F = h\nu_F(\pi n)^{1/2}$. To obtain the influence of the gate voltage on $S$, the term $(1/R)(dR/dE)$ is replaced by $(1/R)(dR/dV_G)(dV_G/dE)$ in the calculation. Solving $dR/dV_G$ and $dV_G/dE$ and bringing the result into (7) yields

$$S = -\frac{2\pi^2}{3} \frac{K_B^2 T}{q} \left( \frac{C_{\text{vas}}(V_{G0})}{q} \right) \sqrt{ \left( \frac{C_{\text{vas}}(V_{G0})}{q} \right)^2 + 4 \left( \frac{n_g^2}{2^2} + \frac{\pi}{6} \left( \frac{K_B^2 T}{n_F} \right)^2 \right) }$$  

(8)

It is obvious that $S$ is controlled by $V_{G0}$ and $T$ in (8). The sign of $S$ is related to the type of carrier. Thus, the carriers induced in the channel are electrons when a positive gate voltage is applied and $S$ is negative. Conversely, $S$ is positive when the carriers in the channel are holes. Fig. 7 shows the variation of $S$ with $V_{G0}$ for different temperatures. At $V_{G0} = 0$ V, when $E_F$ is at the charge neutral point, SLG is more similar to a conductor. As the carrier density is adjusted by the gate voltage, the $E_F$ of SLG is far away from the charge neutral point as $V_{G0}$ increases. $S$ increases significantly with $V_{G0}$ within the low voltage range and then begins to drop slightly in the high voltage range, presenting the expected thermoelectric properties. This phenomenon is consistent with the results in [32]. Moreover, $T$ has a positive correlation with the absolute value of $S$. High $T$ values correspond to high $|S|$. When the majority carrier is a hole, that is, when $V_{G0}$ is negative, $S$ has a positive value. Fig. 8 shows the Seebeck coefficient as a function of $T$ at different hole carrier concentrations. In Fig.8, the sensitivity of $S$ to $T$ at low carrier concentrations is stronger than that at high carrier concentrations because a high gate voltage leads to a high $n_g$ and then acts on $S$. The higher the gate voltage, the larger the $n_g$, and the weaker the regulation of $n_{th}$ on $S$ caused by $T$. For the curve of $n = 0.5 \times 10^{12} \text{ cm}^{-2}$ where $V_{G0} = -5$ V, $S = 156.05 \mu\text{V/K}$ at $T = 300$ K, and $S = 208.07 \mu\text{V/K}$ at $T = 400$ K, whereas $S$ can reach 260.08 $\mu\text{V/K}$ at $T = 500$ K. It can be seen that the elevated temperature can significantly increase $S$ at low $V_{G0}$ values. In addition, taking $T = 300$ K as an example, $S = 134.06 \mu\text{V/K}$ when $n = 1 \times 10^{12} \text{ cm}^{-2}$ and $V_{G0} = -13$ V, and $S = 75.06 \mu\text{V/K}$ when $n = 3 \times 10^{12} \text{ cm}^{-2}$.
and $V_{G0} = -40$ V. Further, $S = 57.39 \mu V/K$ when $n = 5 \times 10^{12}$ cm$^{-2}$ and $V_{G0}$ reaches -67 V. It can be seen that the value of $S$ continues to decrease with the increase of $V_{G0}$. In summary, the regulation of $S$ by appropriate voltage and temperature values is essential to improve the thermoelectric characteristics of SLG.

**B. POWER FACTOR**

The power factor ($Q = S^2 \sigma$) of SLG is also calculated in this paper. As shown in Fig. 9, $Q$ increases first with $V_{G0}$ and then becomes saturated, indicating that an excessively high gate voltage is not useful for improving $Q$. $Q$ begins to stabilize when $V_{G0} > 5$ V at $T = 100 K$. However, $Q$ tends to stabilize when $V_{G0} > 14$ V at $T = 500$. Therefore, the reasonable working range of $V_{G0}$ should be controlled in the range of 5–14 V. In addition, $T$ has a benefit on the improvement of $Q$. However, different semiconductor devices have different production processes, thus their operating temperatures cannot be precisely controlled. For example, given that the operating temperature of a typical device is approximately 300–400 K, it is more convenient to adjust $V_{G0}$ to obtain a higher $Q$. Therefore, appropriate conditions should be selected to obtain the desired results when controlling the thermoelectric properties of graphene.

**C. SEEBECK VOLTAGE**

Based on the definition of the Seebeck effect, $V_S$ is produced when there is a temperature difference between the two ends in the system. One end is controlled to the reference temperature ($T_L$), the other end is the high temperature state ($T_H$), and $T_H$ is set as the variable. $\Delta T = T_H - T_L$. Based on the analysis of the Seebeck coefficient above and the influence of temperature changes on each parameter, the expression of $V_S$ can be written as [36]

$$V_S = -\frac{\pi^2 k_B^2}{3 q \hbar v_F} \left( \frac{C_{co}(V_G - V_0)}{q} \right) \left( \frac{C_{co}(V_G - V_0)}{q} \right) \sqrt{ \frac{C_{co}(V_G - V_0)}{q} } \right) \times \left( T_H - T_L \right)$$

(9)

In Fig. 10, when the temperature difference between the two ends of the SLG is not obvious, $V_S$ is small. However, with the augmentation of $\Delta T$, the increasing velocity of $V_S$ is slightly promoted. Since $V_S$ is proportional to the Seebeck coefficient, the energy of the carrier in the hot end becomes stronger due to a high $T_H$ value, leading to an increase of the difference between the Fermi levels at both ends of the SLG and enhancing the Seebeck effect. In addition, the $V_G = 10$ V curve indicates that a low gate voltage is not beneficial to the generation of $V_S$. In the case of $V_G = 20$ V and $\Delta T = 100$ K, the value of $n$ is approximately $1.1 \times 10^{12}$ cm$^{-2}$, and $V_S$ can reach 12.3 mV. This result is consistent with the results in [35]. With the continued rise of $V_G$, $V_S$ is in a falling state. From the previous analysis, it can be seen that $S$ first increases and then decreases with the increase of $V_{G0}$, and the variation law of $V_S$ with the gate voltage is consistent with that of $S$ with the gate voltage. It can be seen that when SLG is used for energy harvesting, the appropriate gate voltage helps to increase the obtained $V_S$ value.

**IV. CONCLUSION**

In this paper, the influences of $T$ and $V_{G0}$ on the carrier concentration and mobility of SLG are analyzed. The carrier concentration, mobility and the thermoelectric parameters of SLG are combined through the two variables $T$ and $V_{G0}$. The square resistance, the Seebeck coefficient, the power factor and the Seebeck voltage of SLG are calculated, and the mechanism of these thermoelectric parameters under different conditions is analyzed in detail. The results show that $T$ and $V_{G0}$ play different roles in the thermoelectric parameters of SLG at different stages. In addition, the thermoelectric properties of SLG can be regulated by controlling $V_{G0}$ and $T$. The power factor of SLG can also be improved by applying an appropriate gate voltage and temperature. This paper provides a theoretical analysis for a better understanding of the variation characteristics and adjustability of the thermoelectric parameters of SLG.

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ZHI-HAO MA was born in 1995. He received the bachelor’s degree in optical engineering from West Anhui University, Luan, Anhui, in 2016. He is currently pursuing the Master of Engineering degree with the University of Shanghai for Science and Technology. He is committed to research on the thermoelectric characteristics of graphene.

CONG GAO was born in 1994. He received the bachelor’s degree in microelectronics from China Jiliang University, Hangzhou, Zhejiang. He is currently pursuing the Master of Engineering degree in optical engineering with the University of Shanghai for Science and Technology. His research interest includes thermoelectric energy harvesting.

HONG-ZHI JIA was born in 1968. He received the B.S. degree in microelectronics from the Beijing Institute of Technology, Beijing, China, in 1990, and the Ph.D. degree from Xi’an Optical Precision Machinery Research Institute of Chinese Academy of Sciences, Xi’an, China, in 2000. Since 2002, he has been a Faculty Member with the University of Shanghai for Science and Technology, Shanghai, where he is currently a Professor. His current research interests include fiber optical sensors and photodetector.

GUO-RONG SUI was born in 1974. He received the Ph.D. degree in optical engineering from the University of Shanghai for Science and Technology, Shanghai, China, in 2008. Since 2008, he has been a Faculty Member with the University of Shanghai for Science and Technology, Shanghai, where he is currently an Associate Professor. His current research interests include high-speed semiconductor device, integrated optics device, and optical-electrical detection.

XIU-MIN GAO was born in 1978. He received the Ph.D. degree from the Shanghai Institute of Optics and Fine Mechanics (SIOM), in 2006, and joined the University of Shanghai for Science and Technology (USST), in 2017. He is currently a Leader of several research programs, including National Natural Science Foundation of China, National Key Research and Development Plan. He has published more than 60 articles in journals, such as Nature Communications, Scientific Reports, and Applied Optics. He obtained more than 50 patents, and his several research achievements have been industrialized. His current research interests include instruments and meters, smart sensing technology, optical engineering, and precision measurement.

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