

Supplementary Information

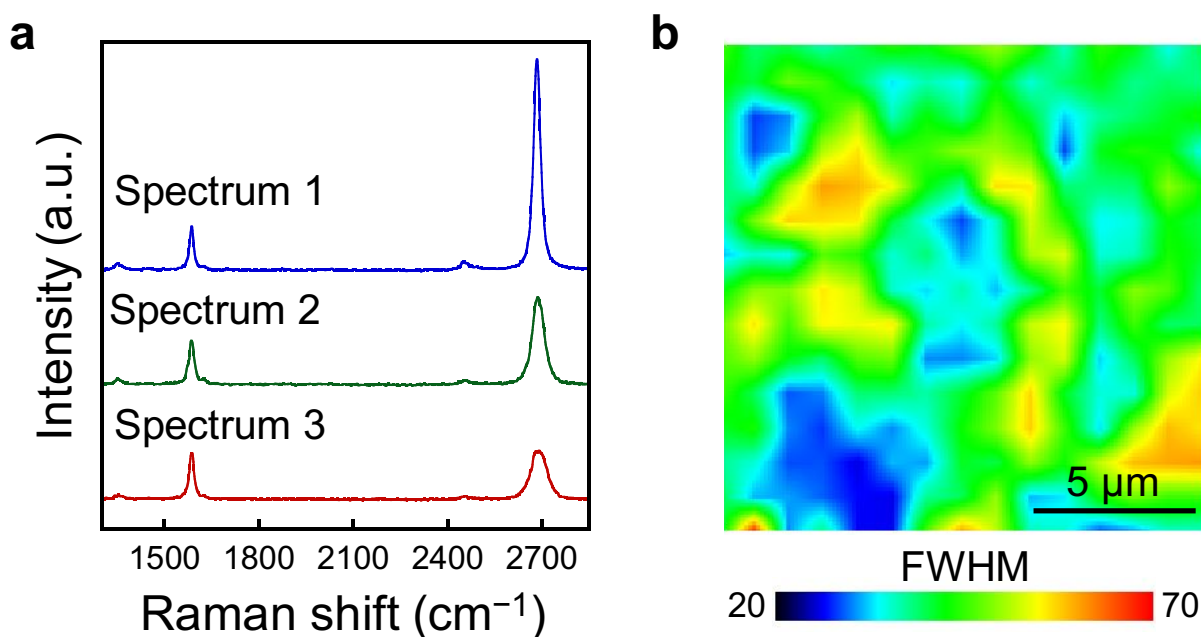
Giant Power Factors in p- and n-type Large-Area Graphene Films on a Flexible Plastic Substrate

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S1. Characterization of CVD-grown graphene films by Raman spectroscopy

We performed Raman spectroscopy measurements to characterize our large-area CVD-grown graphene films. Supplementary Fig. 1a shows three typical Raman spectra obtained from our samples on a glass substrate. As shown in Supplementary Fig. 1a, two main peaks are clearly observed in all of the spectra at $\sim 1580\text{ cm}^{-1}$ and $\sim 2680\text{ cm}^{-1}$, which correspond to the G-band and 2D-band peaks, respectively.¹⁻³ First, the full width at half maximum (FWHM) of the 2D-band peaks in the three spectra are $\sim 26\text{ cm}^{-1}$ (blue, spectrum 1), $\sim 44\text{ cm}^{-1}$ (green, spectrum 2), and $\sim 59\text{ cm}^{-1}$ (red, spectrum 3). Second, the intensity ratios between the 2D and G peaks (2D/G) in the three spectra are calculated as 3.9 (blue, spectrum 1), 2.0 (green, spectrum 2), and 1.1 (red, spectrum 3). The FWHM of the 2D-band peak and the 2D/G ratio of exfoliated single-layer graphene are 24 cm^{-1} and 3.79, respectively.¹⁻³ Therefore, spectrum 1 is assigned as the Raman spectrum corresponding to a region of single-layer graphene. Similarly, spectra 2 and 3 are assigned as Raman spectra corresponding to a region of double-layer graphene and a region of triple-layer graphene, respectively.¹⁻³

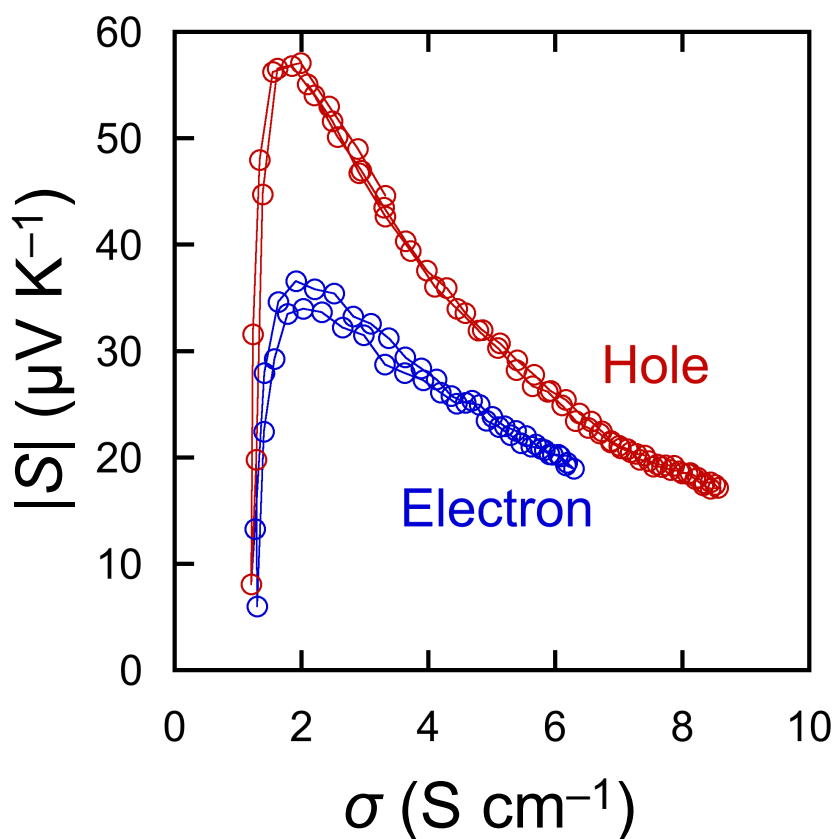
Next, we carried out spatially resolved Raman spectroscopy to estimate the average layer number for our large-area CVD-grown graphene films. Supplementary Fig. 1b shows a color map of the FWHM of the 2D-band peak over a $15\ \mu\text{m} \times 15\ \mu\text{m}$ area. The FWHM values of the 2D-band peak range from $20\ \text{cm}^{-1}$ (blue) to $70\ \text{cm}^{-1}$ (red). Based on Supplementary Fig. 1a, the average values of the 2D/G ratio and FWHM of the 2D-band peak are estimated at 2.0 and $42\ \text{cm}^{-1}$, respectively, which correspond to double-layer graphene; we concluded that the average layer number of our sample is 2.



Supplementary Figure 1. **a** Typical Raman spectra of our graphene samples. The FWHM values of the 2D-band peaks in the three spectra are measured to be $\sim 26\ \text{cm}^{-1}$ (spectrum 1), $\sim 44\ \text{cm}^{-1}$ (spectrum 2), and $\sim 59\ \text{cm}^{-1}$ (spectrum 3). The 2D/G ratios in the three spectra are calculated as 3.9 (spectrum 1), 2.0 (spectrum 2), and 1.1 (spectrum 3). **b** Two-dimensional color mapping of the FWHM of the 2D band over a $15\ \mu\text{m} \times 15\ \mu\text{m}$ area. The scale bar corresponds to $5\ \mu\text{m}$.

S2 The $S - \sigma$ relationship in the CVD-grown large-area graphene films

Supplementary Fig. 2 shows the $|S| - \sigma$ relationship in the EDLT of CVD-grown large-area graphene films on the PET substrate including both forward and backward gate voltage sweeps. As shown in Supplementary Fig. 2, no hysteresis between S and σ is observed, indicating that we have succeeded in measuring the intrinsic thermoelectric properties of our films.



Supplementary Figure 2. The $|S| - \sigma$ relationships for hole and electron regimes in the CVD-grown large-area graphene films.

S3 Metallic $S - \sigma$ relationship explained by the Mott equation

For degenerate or metallic electronic states, S is described by the Mott equation as follows:

$$S = \frac{\pi^2 k_B^2 T}{3e} \frac{\partial \ln \sigma_E(E)}{\partial E} \Big|_{E=E_F} \quad (S1)$$

where k_B is the Boltzmann constant, e is the elementary charge, T is the temperature, $\sigma_E(E)$ is the energy-dependent conductivity function, and E_F is the Fermi energy.⁴ This equation predicts the relation $S \propto \sigma^{-1}$ under the condition that $\partial \sigma_E(E)/\partial E$ is constant.

S4 Estimation of maximum electric power from thermoelectric devices based on 2D materials

Based on our experimental results, we estimate the available maximum electric power from flexible thermoelectric modules. Among various human body areas, a high heat energy flow of $10 \sim 20 \text{ mW cm}^{-2}$ is released from the wrist area to ambient air.^{5,6} If flexible thermoelectric modules generate an output power of $1 \sim 10 \text{ } \mu\text{W}$ from the temperature difference between the human body and ambient air, wearable IoT sensors and/or devices can be operated. However, the typical device structure of thermoelectric modules is not suitable for 2D thermoelectric materials on flexible substrates since the bending direction is vertical to the 2D material film. Very recently, a groundbreaking architectural solution was demonstrated: 3D helical coils formed by 2D thermoelectric films.⁷ It is expected that approximately 10^4 thermoelectric legs can be easily fabricated within an area of 100 cm^2 . Then, under the condition of no contact resistance, the maximum output power ($P_{\max}$) of thermoelectric modules is determined as:

$$P_{\max} = N \frac{(|S_p| + |S_n|)^2 \Delta T^2}{4R_{\text{in}}} \quad (S2)$$

where N is the total number of thermoelectric legs, ΔT is the temperature difference within the thermoelectric modules, and R_{in} is the resistance of the thermoelectric legs.⁵ For a simple estimation of P_{max} using our optimized graphene films, we introduce three assumptions: (i) the ratio between the length (L) and the width (W) of both p- and n-type graphene films is fixed at $L/W = 5$; (ii) the temperature difference (ΔT) induced between the human body ($\sim 37^\circ\text{C}$) and ambient air ($\sim 22^\circ\text{C}$) is 5 K; and (iii) the area of the thermoelectric module is 100 cm^2 . Under these assumptions, although 100 cm^2 corresponds to 10^4 thermoelectric legs, the calculated P_{max} reaches only $0.09\text{ }\mu\text{W}$ due to the small volume of the graphene films. Therefore, the stacking of thermoelectric layers is significantly effective. If optimized large-area graphene films can be prepared on commercially affordable thin ($\sim 1\text{ }\mu\text{m}$) plastic substrates, the total film thickness of 100 stacked layers corresponds to $100\text{ }\mu\text{m}$, which is almost half that of the current PET substrate. Following this argument, if we can fabricate thermoelectric modules of $10 \sim 100\text{ cm}^2$ using these stacked thermoelectric films, the output power reaches approximately $1 \sim 10\text{ }\mu\text{W}$.

References

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