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Modulation of Thermoelectric Performance by Using Electrolyte Gating Method

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Abstract

We investigate the carrier density dependence of electrical conductivity and Seebeck coefficient of large-area MoS_2 and WSe_2 monolayers, few-layer graphene and organic polymer DPPT-TT using the electrolyte gating method. The polarity of the thermoelectric properties changes as the majority carrier density switches from electron to hole. By the wide and contentious control of carrier density, we can easily obtain the maximum power factor and optimum carrier density in the same sample.

1. Introduction

Thermoelectric energy conversion is one of the key techniques for energy harvesting devices to convert waste heat into electric energy. Particularly, Seebeck coefficient S, which is the proportional constant to the thermoelectromotive force against a temperature difference, is a significant factor in thermoelectric materials. For efficient use of waste heat from curved surface, such as piping and body, it is necessary to investigate thermoelectric performance in flexible materials. Therefore, we selected the promising flexible materials, such as transition metal dichalcogenide (TMDC) monolayers (MoS_2 and WSe_2), few-layer (FL) graphene and organic polymer (DPPT-TT). Especially, Hicks and Dresselhaus theoretically predicted thermoelectric properties can be enhanced by quantum effects in low-dimensional materials in 1993 [1]. However, since Seebeck coefficient and power factor generally depend on carrier density, it is important to obtain the carrier density dependence of thermoelectric properties. To achieve this objective, we focus on the electric double layer transistors (EDLTs), in which the gate dielectric layer are replaced with electrolytes. Importantly, owing to the huge specific capacitance of EDL, we can tune the carrier density widely and continuously, which results in the control of thermoelectric properties easily.

2. Research Purpose

We have already succeeded in fabricating EDLT and measuring thermoelectric properties in large-area TMDC monolayers [2]. In this work, we expanded the target materials (FL graphene and organic polymer) and investigated the thermoelectric performances in these samples. In addition, we compare the thermoelectric performance between TMDC monolayers, FL graphene and organic polymer.

3. Experimental Section

Polycrystalline TMDC monolayer films were synthesized onto sapphire substrates by a chemical vapor deposition method. Large-area FL graphene films were also produced on Cu foils (33 μm thick) by the surface wave plasma CVD method. After that, the graphene films were transferred onto polyethylene terephthalate substrates (188 μm thick). Organic polymer layers, such as DPPT-TT, were prepared onto glass substrate by a spin-coating method. Au/Ni (30 ~ 80 nm/ 3 nm) electrodes were thermally deposited as the source, drain and gate electrode. Importantly, we introduced a long channel length of 0.4 mm and width of 2 mm, which are crucial to induce a clear temperature gradient for reliable thermoelectric measurements. As the gate dielectric, we selected an ion gel, which was made from an ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]) and an organic polymer Poly(vinylidene fluoride-co-hexafluoropropylene) (P(VDF-HFP)). The gate dielectric layer was prepared by spin-coating method.

Measurements of the gate voltage dependence of Seebeck coefficient and electrical conductivity were performed under nitrogen-filled glove box. The schematic view of thermoelectric measurement system is shown in Fig. 1. The measurement procedure are as follows. The temperature gradient was established by two Peltier elements. The temperature difference ΔT increased over 1 K. In each gate voltage V_G, the temperature difference ΔT and thermoelectromotive force ΔV were measured by two K-type thermocouples and voltage meter, respectively. The Seebeck coefficients were defined as the slope of the $\Delta V - \Delta T$ profiles in each gate voltage V_G.

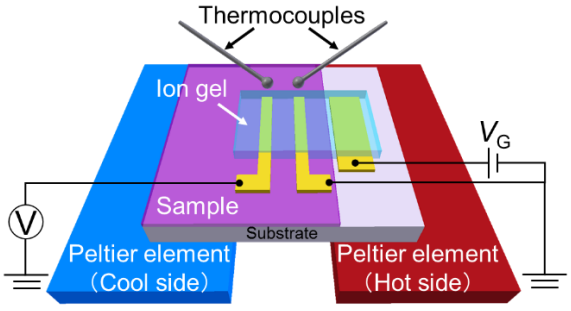

Fig. 1. Schematic view of thermoelectric measurement system

4. Results

Figure 2(a) shows the gate voltage V_G dependence of electrical conductivity σ in FL graphene-EDLT sample. In this work, the thickness of film is assumed to be 0.6 nm. As shown in Fig. 2(a), we observed ambipolar curve. Since the sign of Seebeck coefficient is the same as the polarity of majority carrier, we can determine which carrier is dominant by thermoelectric measurement. Figure 2(b) shows the gate voltage V_G dependence of Seebeck coefficient S in FL graphene-EDLT sample. As shown in Fig. 2(b), the sign of Seebeck coefficient changes as the gate voltage V_G is tuned. Especially, the inflection point of Seebeck coefficient correspond with that of electrical conductivity. Therefore, the sign of majority carrier switches from hole to electron on the boundary of near the gate voltage of 0.2 V. This is similar to previous reports using micrometer-scale bilayer graphene samples [3]. In addition, power factor PF (= $S^2\sigma$) is also important factor to evaluate the output power from a temperature difference. Figure 2(c) shows the gate voltage V_G dependence of power factor (= $S^2\sigma$) in FL graphene-EDLT sample. In both p- and n-type region, optimized power factor is shown. From this result, we can extract the optimum hole and electron density in the same FL graphene sample. As the summary, we achieved the modulation of thermoelectric performance by wide and contentious controlling carrier density. Also in the other sample, such as TMDC monolayers and organic polymer, we achieved the modulation of thermoelectric properties by electrolyte gating method.

5. Conclusions

We fabricated electric double layer transistors (EDLTs) using polycrystalline TMDC monolayer, FL graphene and organic polymer films. Using the huge specific capacitance originated from electrolyte, we succeeded in controlling the thermoelectric properties continuously in the above samples. Especially, we revealed the optimized power factor PF (= $S^2\sigma$) and the optimum carrier density in the same sample by our technique. Therefore, our EDLT method may promote extraction of maximum thermoelectric performance in thermoelectric materials. In the conference, we will discuss the details of electrical conductivity dependence of Seebeck coefficient and power factor PF and compare the thermoelectric performances of our samples.

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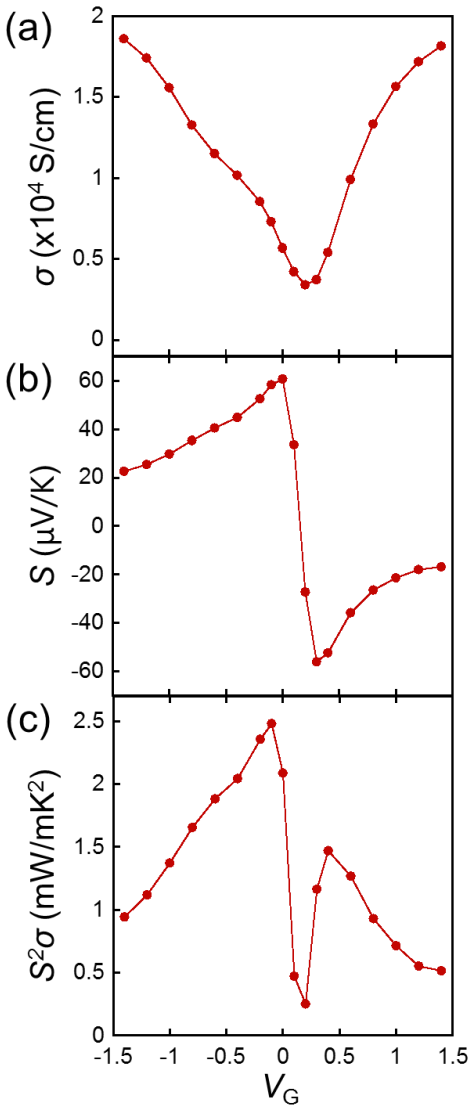

Fig. 2. Gate voltage V_G dependence of electrical conductivity σ (the thickness of film is assumed to be 0.6 nm), Seebeck coefficient S and Power factor PF (= $S^2\sigma$) in few-layer graphene-EDLT sample.