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Correcting for contact geometry in Seebeck coefficient measurements of thin film devices

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ABSTRACT

Driven by promising recent results, there has been a revived interest in the thermoelectric properties of organic (semi)conductors. Concomitantly, there is a need to probe the Seebeck coefficient S of modestly conducting materials in thin film geometry. Here we show that geometries that seem desirable from a signal-to-noise perspective may induce systematic errors in the measured value of S , S_m , by a factor 3 or more. The enhancement of S_m by the device geometry is related to competing conduction paths outside the region between the electrodes. We derive a universal scaling curve that allows correcting for this and show that structuring the semiconductor is not needed for the optimal electrode configuration, being a set of narrow, parallel strips.

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1. Introduction

The use of organic materials for thermoelectric applications [1–16] has rapidly gained popularity in the past few years. Compared to e.g. inorganic materials, organic materials offer relatively low thermal conductivity [17] and good processability and scalability [2,11]. In combination with the absence of rare elements the latter properties make organic materials potentially relevant for large-area harvesting of (waste) heat. The thermal conductivity is however not the only parameter that defines the quality of a material to be used as thermoelectric generator. This quality is expressed by the thermoelectric figure of merit $ZT = S^2 \sigma T / K$ [18], where S is the Seebeck coefficient, σ the electrical conductivity, T the absolute temperature, and K the thermal conductivity. Recent work [2,3,19] on materials based on poly(3,4-ethylenedioxythiophene) (PEDOT) has shown the potential of doped organic semiconductors

for thermoelectric applications and the reported record value of ZT in organic materials has increased up to 0.42 [3], approaching the record value in inorganic materials, $ZT = 1.4$, in bismuth antimony telluride.

With the field of organic thermoelectrics gaining momentum, it is highly desirable that consistent methodologies to determine ZT are employed. To determine the figure of merit of a material, S , σ and K need to be determined. In this paper we focus on the measurement of S , and show that, when working in thin-film geometry, Seebeck measurements are particularly prone to suffer from systematic and substantial overestimation of S . It is the purpose of this paper to assist in the design and interpretation of Seebeck measurements on thin organic films.

The Seebeck coefficient is the ratio of the thermovoltage and its driving force, the temperature difference:

$$S = \Delta V / \Delta T. \quad (1)$$

The magnitude of S is related to the temperature dependence of the Fermi level. An important consequence of this equation is that a small temperature gradient does not necessarily lead to a small thermovoltage as it is its

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integral, the temperature difference, between the two electrodes that matters. This is in contrast to electrical currents that are driven by voltage gradients, i.e. electric fields. Ideally, voltage probes are therefore infinitely small or otherwise forced to have a constant temperature over their entire area. When not, the thermovoltage becomes ill-defined and erroneous values of S are measured. Triggered by the differences in electrode geometries in recent reports [2–4,9–14] and our failure to reproduce some of them we here address the errors that can be made in measuring S of thin films with experimentally practical device geometries.

In practice, measurement of S in organic (semi)conductor thin films, that are typically of low to modest conductivity, demands the use of metal electrodes of finite size to monitor the thermovoltage. The reason is that the device impedance, as probed between the electrodes, needs to be small compared to the input impedance of the measurement setup. In literature different electrode geometries are encountered, e.g. long narrow electrodes [4,19–20], roughly square electrodes [3], point-like contacts [9,14], and more complex structures [2,11]. One of the considerations when choosing the contact geometry is the conductivity of the studied material. To improve the signal-to-noise ratio short channels with a large cross-sectional area are preferred. Therefore point contacts are often not feasible to measure Seebeck coefficients in thin films. For an ideal Seebeck coefficient measurement with finite-sized electrodes current flow is only allowed within the channel between the two electrodes as is the case in Ref. [2]. In other cases [3,4,10,19–21], the electrodes are placed on top of or below a larger semiconducting film, enabling additional current flow outside the channel. When a linear temperature gradient is applied in a region that includes the electrodes, as for practical reasons is commonly done, this electrode layout possibly distorts the measured value of S : Fig. 1 shows a typical experimental geometry in which

the thermovoltage ΔV_{23} is smaller than ΔV_{14} as ΔT_{23} is smaller than ΔT_{14} . The intuitive assumption that ΔV_{23} will dominate the measurement need not be correct, as will be quantified below by numerical and analytical calculations and experiments.

By means of numerical modeling we show that an inappropriate choice in device geometry can lead to a measured Seebeck coefficient, S_m , that is larger than the actual Seebeck coefficient, S_0 , of the studied material by up to a factor 3 in case square-shaped electrodes are used. This enhancement is driven by the larger temperature difference outside the channel, resulting in additional conduction pathways, enhancing the thermovoltage. The difference between S_m and S_0 resulting from this process can be reduced significantly by removal of the active material outside the channel; the use of a set of parallel, narrow line-shaped electrodes; or simply by determination and compensation of the error in S_m , i.e. $S_m - S_0$. The latter is facilitated by the fact that the geometry-induced error in S follows a universal scaling behavior, so the results presented below can be used to correct any experiment conducted with the layout sketched in Fig. 1. The numerical calculations are confirmed in relevant limits by intuitive analytical derivations based on Ohm's law, which prove that the described effect must be present – in fact, finding an independence on geometry should set off alarm bells. Experiments have shown that partial patterning of the active layer outside of the channel indeed results in a better approximation of S_0 by S_m .

2. Materials and methods

2.1. Computational details

A top-view of the device geometry considered in the numerical simulations is shown in Fig. 1. The model is used to calculate the thermovoltage which results from a linearly distributed temperature difference $\Delta T = T_2 - T_1$, which is applied across the complete device. Carrier transport is calculated on a 2D rectangular grid by forward integration in time of the Boltzmann transport equation. Poisson's equation is used to calculate the potential after each time step. The electrodes have a fixed potential and hole injection into its neighboring cells is described by setting a fixed, large density of free holes at the contact edges, which is a commonly accepted way to mimic Ohmic contacts. For simplicity, a delta-shaped density of states was used, which is known to give rise to large Seebeck coefficients, $S_0 \approx 340 \mu\text{V/K}$ for the used parameters.[22] However, since we are interested in the relative influence of the device geometry, the magnitude of S_0 is irrelevant. To model a doped system like PEDOT:PSS, a fixed density of negatively charged and immobile dopants ($n_{\text{dopant}} = 1 \cdot 10^{25} \text{ m}^{-3}$) is added to the semiconductor, compensated by the same density of mobile holes. The dimensions of the semiconductor surrounding the contacts were made sufficiently large that further increases did not affect the thermovoltage anymore. More details on the numerical model can be found in the [Supplementary Information Part A](#). Importantly, since relative errors are reported below, the

Fig. 1. Schematic layout of the modeled device geometry for Seebeck measurements. Color gradients reflect the temperature profile in the semiconductor.

material-specific parameters in the model are of no effect to our conclusions.

2.2. Experimental details

Devices were prepared on pre-cleaned 1 mm thick glass substrates. Electrodes were deposited by thermal evaporation of Cr (5 nm) and Au (100 nm) through a shadow mask in vacuum. Materials to be characterized were spin-cast on top of the substrate. Either one of two variants of doped poly(3,4-ethylenedioxythiophene) (PEDOT) stabilized by poly(styrenesulfonate) (PSS) were characterized: Clevios 4083 and Clevios PH1000 from H.C. Starck. The latter was mixed with 5 vol% DMSO prior to spin coating. After spin coating the layers were dried on a hotplate at 120 °C for 10 min. The final layer thickness of the films was ≈ 60 nm. Sample preparation was performed in air.

Measurement of S_m and σ was performed under nitrogen atmosphere ($H_2O < 1$ ppm and $O_2 < 1$ ppm) in a glovebox. A Keithley source measure unit 2636 was used to measure both thermovoltages and conductivities. Before measurement samples were annealed again on a hotplate in the glovebox at 120 °C for 10 min. The substrate was positioned on two Peltier elements in such a way that a temperature gradient can be established across the substrate. Note that the thermal conductance of the ≈ 100 nm thick electrodes is negligible compared to the thermal conductance of the 1.5 mm thick glass substrate. Consequently, a linear temperature profile can be assumed across the glass substrate between the Peltier elements. Two silicon diodes were used to measure the temperature on each side of the sample. The electrodes were contacted by Au probe needles. The thermovoltage ΔV at a given temperature gradient ΔT was determined by measurement of the zero crossing of the I - V characteristic. S_m was then determined by fitting the measured ΔV over a range of ΔT with Eq. (1).

3. Results and discussion

3.1. Numerical calculation of the error in the measured Seebeck coefficient

To study different device geometries by numerical calculations, we varied the aspect ratio of the channel,

$$r_c = \frac{W_e}{L_c}, \quad (2)$$

and the aspect ratio of the electrodes,

$$r_e = \frac{W_e}{L_e}. \quad (3)$$

Eqs. (2) and (3) reflect that the entire problem is scale invariant. The device Seebeck coefficient, S_m , is calculated from Eq. (1) under the (incorrect) assumption that the obtained thermovoltage represents ΔV_{23} (see Fig. 1) and, is plotted in Fig. 2a for different aspect ratios.

The results show that small relative errors result when the channel length L_c is much longer than the electrode length L_e , i.e. when $r_c/r_e \ll 1$. In this limit the relative

difference between T_{14} and T_{23} , and hence between V_{14} and V_{23} becomes vanishingly small. In practice this easily leads to such an increase in device impedance that noise and sensitivity become an issue.

For larger channel aspect ratios, the modeling reveals a significant dependence of the measured Seebeck coefficient on the electrode aspect ratio. Large aspect ratio electrodes, i.e. wide and short, (triangles in Fig. 2) clearly result in the most accurate approximation of S_0 , whereas square-shaped electrodes (squares in Fig. 2) easily lead to an overestimation of S_0 by a factor >3 .

3.2. Analytical limits for the error in the measured Seebeck coefficient

The relative error in the measured Seebeck coefficient in Fig. 2a is observed to have two limits related to the channel aspect ratio: for $r_c \gg 1$, the relative error is roughly constant, whereas for $r_c \ll 1$ the error is observed to decrease linearly. In the following paragraphs intuitive analytical derivations are given for both these limits to gain understanding regarding the origin of the error in the measured Seebeck coefficient. This derivation is based on Ohm's law.

The thermovoltage that is measured ($V_{th,m}$), can intuitively be separated into two components: one related to the actual channel potential V_{23} and a spurious one related to the potential V_{14} that drops in the regions outside and far away from the channel. To determine the influence of these different regions, the conductance associated with each contribution should be used as weighting factor:

$$V_{th,m} \sim \frac{V_{23} \cdot G_{23} + V_{14} \cdot G_{14}}{G_{23} + G_{14}}, \quad (4)$$

where $G_{23} = \sigma_{PEDOT} \frac{d \cdot W_e}{L_c}$ is the conductance of the PEDOT in the channel between the electrodes and G_{14} is related to the conductance of the PEDOT outside the channel. d_{PEDOT} is the thickness of the layer which is of no further importance.

First we consider the limit $r_c \gg 1$. Suppose the geometry of the electrodes is constant ($r_e = \text{constant}$) and that r_c increases by a reduction of L_c . In the limit $r_c \gg 1$, where $r_e = \text{constant}$, both V_{14} and G_{14} can be considered constant as well. G_{23} however increases with decreasing L_c (and increasing r_c) as $1/L_c$, whereas V_{23} decreases linearly. Eq. (4) then predicts that the thermovoltage should decrease linearly as only the denominator increases linearly ($G_{23} \gg G_{14}$):

$$V_{th,m} \sim V_{23} + V_{14} \frac{G_{14}}{G_{23}}. \quad (5)$$

The relative error in the thermovoltage then becomes:

$$\frac{V_{th,m}}{V_{23}} - 1 \sim \frac{V_{23}}{V_{23}} + \frac{V_{14}}{V_{23}} \frac{G_{14}}{G_{23}} - 1 = \frac{V_{14}}{V_{23}} \frac{G_{14}}{G_{23}}. \quad (6)$$

As V_{14} and G_{14} can be considered constant for decreasing L_c in the limit $r_c \gg 1$, the simultaneously increasing G_{23} and decreasing V_{23} compensate each other, making the relative error constant. This is in agreement with the constant region in Fig. 1b for $r_c \gg 1$. The saturation value of about $2/r_e$ can be handwavingly rationalized by a simple

Fig. 2. (a) Relative error in measured Seebeck coefficient for different channel and electrode aspect ratios. The error in the Seebeck coefficient of the device S_m is relative to the true Seebeck coefficient of the modeled semiconductor S_0 and (b) the relative error in the measured Seebeck coefficient multiplied by the electrode aspect ratio r_e reveals a universal curve. The solid black lines indicate the analytical limits Eqs. (6) and (10) discussed in the text.

square-counting argument as described in the [Supplementary Information Part B](#). An important consequence of this constant relative error is that differential probes, in which S_m is obtained as a function of channel length do not avoid this geometry-related problem as long as the channel remains short.

In the other limit, $r_c \ll 1$, again Eq. (4) is used as starting point. Now V_{14} can be expressed as:

$$V_{14} = V_{23} + \Delta, \quad (7)$$

where

$$\Delta = 2V_{23} \frac{L_e}{L_c} = 2V_{23} \frac{r_c}{r_e}. \quad (8)$$

Combination of Eqs. (4), (7), and (8) gives the relative error in the thermovoltage, which is linearly related to the Seebeck coefficient:

$$\frac{V_{th,m}}{V_{23}} - 1 = 2 \frac{r_c}{r_e} \frac{G_{14}}{G_{23} + G_{14}}. \quad (9)$$

For small r_c , the points 1 and 2 as well as 3 and 4 in Fig. 1 collapse, resulting in $G_{14} = G_{23}$. Hence Eq. (9) reduces to

$$\frac{V_{th,m}}{V_{23}} - 1 = \frac{r_c}{r_e}, \quad (10)$$

which is linear in r_c for constant r_e , in agreement with the numerically calculated relative error in the regime $r_c \ll 1$ shown in Fig. 2a. In addition we note that the relative error multiplied by the electrode aspect ratio r_e only depends on the channel aspect ratio as is shown in Fig. 2b.

3.3. Experimental verification of geometry influence

To verify the modeled results experimentally, two Seebeck coefficient measurements on PEDOT:PSS (Clevios 4083) with different channel and electrode aspect ratios were performed as shown in Fig. 3a and b. For the aspect ratios $r_c = 7$ and $r_e = 7$ the measured Seebeck coefficient

was found to be $5 \pm 2 \mu\text{V/K}$. For the aspect ratios $r_c = 37.5$ and $r_e = 0.75$ the measured Seebeck coefficient was found to be $22 \pm 4 \mu\text{V/K}$. This is in quantitative agreement with the numerical model that predicts errors (i.e. S_m/S_0) of respectively ~ 1.25 and ~ 3.8 for these geometries. Compensating for the geometry, the values of S_0 are $4 \pm 2 \mu\text{V/K}$ and $6 \pm 1 \mu\text{V/K}$ for $r_e = 7$ and 0.75 , respectively.

Similar measurements on PEDOT:PSS (Clevios PH1000) are shown in Fig. 3d. In addition, the distance between the Au probes contacting the pads was varied to eliminate any probe-related effects. Altering the inter-probe distance clearly has an effect on the resultant value of S_m , which is determined by the temperature difference across the channel and the measured thermovoltage using Eq. (1). Dashed lines are added to indicate the trend of this behavior. At the limit of zero inter-probe distance spurious thermoelectric effects due to the probe-contact interfaces vanish, leaving purely the measured Seebeck coefficient of the PEDOT. Again, the use of electrodes with $r_e = 0.75$ results in a relatively large error in S_m of approximately a factor 4 compared to a device with $r_e = 7$. The value of $S_m = 30 \pm 3 \mu\text{V/K}$ for $r_e = 0.75$ is similar to the values obtained in Ref. [3] for a similar device geometry with the same PEDOT:PSS material (Clevios PH1000 from H.C. Starck). Compensating for the geometry, the calculated values of S_0 are $6 \pm 2 \mu\text{V/K}$ and $8 \pm 1 \mu\text{V/K}$ for $r_e = 7$ and 0.75 , respectively. The resistivity values of the PEDOT was determined to be 7.5 and $5.2 \mu\Omega/\text{m}$ for the rectangular and square electrode geometry, respectively, which indicates that the spin-coated films are similar regarding electrical conductivity despite a difference in electrode geometry. The I - V characteristics from which the thermovoltage was determined were furthermore linear as shown in Fig. C.1 in the [Supplementary Information Part C](#). This indicates that the electrodes are ohmic contacts and the results are therefore not affected by temperature dependent injection. The relatively large error bars shown in Fig. 3d are related to the noise in the I - V characteristics shown in Fig. C.1. To reduce the error,

Fig. 3. Schematic representation of (a) and (b) unstructured devices with an electrode geometry of (a) $r_e = r_c = 7$ and (b) $r_e = 0.75$ and $r_c = 15$ and a schematic representation of (c) a structured device with an electrode geometry of $r_e = 0.75$ and $r_c = 15$. Structuring of PEDOT on the glass substrate is indicated by the different gray tints. The dotted lines indicate the position of the Peltier elements, and (d) experimentally obtained Seebeck coefficient in PEDOT:PSS PH1000 from H.C. Starck for different device geometries as indicated in the legend and shown in (a)–(c). The dashed lines indicate the effect of the Au probes with varying inter-probe distance as indicated by the red dots in (a)–(c). Au contact pads were used. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the measured Seebeck coefficient was determined from linear fits of measurements over relatively large voltage intervals of 2 mV.

3.4. Other considerations

In addition to the error in the S_m due to the presence of PEDOT next to the electrodes, also the presence of PEDOT underneath or on top of the metal electrodes results, for the same reasons, in an error in S_m . For the geometry $r_c/r_e = L_e/L_c = 24.5$, e.g. a device with channel length of 50 μm and electrode length of 1.2 mm, the relative error in S_m was calculated to be 0.18–0.40 for PEDOT layer thicknesses of 60–180 nm respectively. As the PEDOT under or on top of the electrodes forms an additional, parallel conduction path from electrode to electrode, this relative error may be added to the relative error discussed in Fig. 2.

Also the effect of partial removal of PEDOT next to the electrodes was calculated. Referring to Fig. 1, PEDOT was removed starting at a distance $0.4 \times W_e$ above and below the electrodes. The relative error in S_m was reduced from ~ 2 to 0.6 and 0.5 for $r_c = 20$ and 10, respectively. Experimentally a similar error reduction is observed after partial removal of the PEDOT to ~ 0.5 mm as shown in Fig. 3c, i.e. equivalent to $\sim 0.33 \times W_e$, above and below the electrodes as shown by the blue square in Fig. 3d.

Next to the overestimation of S_0 by a non-optimal choice in device geometry, also a difference in S_0 between the contact pads and the contact probes can give rise to spurious thermovoltages. Also a difference in S_0 is possible between, e.g., a thin amorphous film (electrode) and a massive rod (probe) of the same metal. The slope of the

orange dashed line in Fig. 3d indicates a difference in S_0 between the ~ 100 nm Au contact pads and the Au contact probes. The error margins on the data in Fig. 3d preclude making a meaningful estimate for the difference in Seebeck coefficient between probes and pads from the slope of the dashed lines.

Besides the measured Seebeck coefficient also the measured conductivity, σ_m , depends on the shape of the electrodes and the length of the channel. Note that this is the case when σ_m is calculated from the measured current, the applied bias voltage, the channel length and the channel width without taking into account the current outside

Fig. 4. Relative error in measured conductivity for different channel and electrode aspect ratios.

the channel. The results are shown in Fig. 4. The relative error in σ_m compared to the actual conductivity, σ_0 , is found to be weakly dependent of the electrode aspect ratio r_e . Square-shaped electrodes ($r_e = 1$) result in a slightly larger error as opposed to longitudinal electrodes placed parallel of each other ($r_e > 1$). The channel aspect ratio r_c has, however, a stronger influence on the relative error. Placement of the electrodes further apart (i.e. a reduction of r_c) leads to an increase in the relative error. The origin of this is that the conductance of the channel between the electrodes approaches the conductance of pathways outside the channel, which leads to a larger measured conductivity.

The reason why the relative errors in σ_m , Fig. 4, are much smaller than those in S_m , Fig. 2, is quite crucial. In both situations spurious contributions arise from current pathways outside the intended channel, and their relative weight, that is set by the conductance associated with those pathways, is equal in both cases. In the case of Fig. 4 the error is a current that is driven by a constant (applied) voltage over an increasing pathway length. However, in the case of Fig. 2 the driving thermovoltage increases with increasing pathway, c.f. Eq. (1), because of the linear temperature profile.

The results discussed in this paper prove that a detailed specification of the used device geometry is required when reporting the measurement of Seebeck coefficients in films. In for example the references [10,20,23,24] no or an unclear description is given of the tested device. It should be noted that the results shown in this paper are also relevant for devices with non-rectangular electrode geometry, such as finite-sized point contacts. For point-like contacts r_e equals 1, which is not optimal regarding the relative error in S_m (see squares in Fig. 2a). To reduce the measurement error (see Fig. 2a) a low value of r_c must be chosen, which is achieved by placing the contacts far away from each other. The cost is, however, a worse signal-to-noise ratio.

4. Conclusions

Overall we have shown that in thin film devices where the active layer is not structured the contact geometry matters a lot to the experimentally obtained Seebeck coefficient, and to a lesser extent to the obtained conductivity. This conclusion is substantiated by numerical and analytical modeling, and experiments. In terms of signal to noise

and an accurate determination of the actual Seebeck coefficient, the optimal configuration is a set of parallel, narrow line-shaped electrodes. We found a universal dependence of the relative error on device geometry which allows one to *a posteriori* correct measurement results.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.orgel.2014.06.018>.

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