

Direct Measurement of the Effective Electronic Temperature in Organic Semiconductors

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Organic semiconductors show complex phenomena due to their high energetic disorder. A striking example is the possibility of an increased effective temperature T_{eff} of the charge carrier distribution relative to the lattice temperature, which results from the slow charge carrier relaxation after excitation, either by high electric field or photon absorption. The increased effective temperature has been linked to conductivity enhancements and performance increases in actual devices, but a direct observation has been lacking. Here, we utilize nanoscopic tree-terminal devices to measure the Seebeck voltage arising in a doped organic polymer semiconductor due to a field-driven enhancement of the effective electronic temperature, providing direct proof of the existence of T_{eff} . The results agree quantitatively with numerical predictions by a kinetic Monte Carlo model. The findings not only provide fundamental understanding but also indicate an avenue toward low-loss thermoelectric devices.

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While it is widely accepted that disorder is a crucial characteristic of virtually all organic semiconductors, there is still debate about its exact influence on charge transport. [1–4] This becomes apparent in the discussion about field behavior. Initially, the effect of conductivity increasing with electric field strength was described in terms of Pool-Frenkel behavior of the type $\sigma \propto \exp(\sqrt{F})$ [5]. It is argued, however, that this description is phenomenological at best, specifically at high fields, and alternative approaches have been proposed [1,6–8]. A common and physically elegant approach focuses on the role of slowly relaxing carriers, which allow the energy distribution to be driven out of equilibrium by modest fields of order 10^7 V/m, which are not uncommon in typical organic thin film devices [9]. The effect of the applied field is then described as a modified, increased temperature of the charge carrier distribution. This concept is illustrated for electrons in Fig. 1, but holds, *mutatis mutandis*, for holes as well. As the charges hop under the influence of an electric field, they gain additional energy due to the drop in potential. If carrier relaxation is sufficiently slow, the distribution of occupied states shifts upward as if having an elevated temperature, called the effective temperature (T_{eff}), relative to the unchanged lattice temperature. From simulations it was phenomenologically described as

$$T_{\text{eff}} = \left[T_{\text{lat}}^\beta + \left(\gamma \frac{e|F|\alpha}{k_b} \right)^\beta \right]^\frac{1}{\beta}, \quad (1)$$

with the lattice temperature T_{lat} , the electric field strength F , the localization length α , and the constants $\beta > 1$ and $\gamma = 0.3\text{--}0.9$. The constants e and k_b represent the elementary charge and the Boltzmann constant, respectively [10–13]. Conductivity is then obtained by replacing the lattice temperature with the effective temperature in the underlying Gaussian disorder model, i.e., $\sigma(T_{\text{lat}}, F) = \sigma(T_{\text{eff}}(T_{\text{lat}}, F), F = 0)$ [1]. While this concept is well-tested

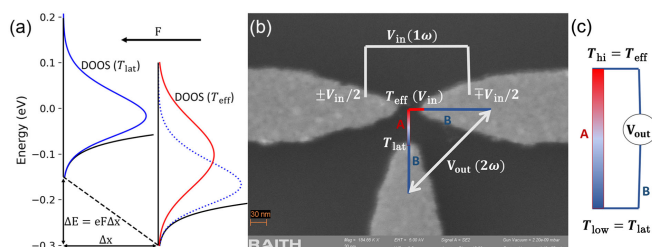


FIG. 1. (a) Schematic illustration of the shift of the electron density of occupied states (DOOS, colored lines) under the application of an electric field F . The density of states (DOS, gray and black lines), shifts with the potential as charges travel to a site located at a distance Δx . Because of the slow carrier relaxation, the zero-field lattice temperature distribution (blue) is replaced by the effective temperature distribution (red). (b) Schematic depiction of the experiment, showing the input and output circuit overlaid on an electron microscopy picture of the center tips. For the actual experiment, the entire structure is covered in $F_4\text{TCNQ}$ -doped P3HT. The application of a voltage V_{in} at frequency ω leads to effective temperature $\propto V_{\text{in}}^2$ and hence a Seebeck voltage V_{out} at 2ω . (c) The equivalent Seebeck circuit with materials A and B representing the organic and gold layers, respectively.

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in simulations of single materials, the exact expression lacks a physical justification. In addition, on the basis of master equation simulations, the concept has been argued to be inapplicable in host-guest systems [14]. Surprisingly little experimental work is devoted to putting the concept on more solid footing, especially for organic semiconductors [7]. In part, this may be due to the combination of the (large) energetic disorder and exciton binding energy that preclude the use of conventional optical approaches to measure electronic temperatures [9,15].

The previous experiments known to us can be separated into two approaches, which have both been pursued by Nebel *et al.* in amorphous hydrogenated silicon. The first directly measures temperature and field scaling of conductivity and checks for equivalence when applying the effective temperature model [16]. The second relies on photoconductivity experiments performed in p-i-n junctions [17]. Here, the equivalent scaling with field and temperature of the initial current decay after photoexcitation and also the drift mobility are analyzed. For Nebel *et al.* all approaches yield good fits with slightly varying localization lengths between 5 and 10 Å. Follow-up photoconductivity experiments in the same material are mixed, with some studies finding reasonable agreement [18,19], while Gu *et al.* [20] cannot fully describe their results in terms of the model, attributing this to improved photoexcitation conditions. Later studies on amorphous carbon compounds also utilize modified versions of the effective temperature concept treating $\gamma\alpha$ as a field-dependent variable [21]. These existing experiments are mostly indirect in the sense that they demonstrate only the equivalence between temperature and field scaling of conductivity or current decay when expressing field in terms of temperature as per the equation above, which formally does not prove the existence of a (more or less) thermal distribution of charge carriers characterized by an enhanced temperature [16].

In line with the above, the possibility of a raised effective temperature is rarely considered in device design, despite potentially substantial implications. As recently demonstrated, the efficiency of organic solar cells may be significantly improved by their ability to utilize a similar distribution shift due to the excess energy of incoming photons [15,22]. Here, the idea is that charges driven out of equilibrium generate an (additional) Seebeck voltage, enhancing device performance. In case it would be possible to couple general excess energy, for instance in the form of stray electric fields, to the charge carrier distribution only, rather than to the full device including the lattice, highly efficient field-driven thermoelectric devices could be realized [23,24]. The reason is that raising only the charge carrier temperature leads to a device that is not, or less, affected by the lattice thermal conductivity, which represents a significant loss channel in current thermoelectric devices. This can be illustrated by the figure of merit for thermoelectrics $ZT = S^2\sigma_e T/(\kappa_e + \kappa_l)$

[25,26]. Using the Wiedemann-Franz law as lowest order approximation for the electronic part of the thermal conductivity $\kappa_e = L_0 T \sigma_e$ with σ_e the electrical conductivity and $L_0 = (\pi^2/3)(k_B/q)^2 \cong 2.44 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$ the Lorenz number, gives, for negligible lattice thermal conductivity κ_l , $ZT = S^2/L_0$ [27]. For reasonable values of the Seebeck coefficient $S > 200 \text{ } \mu\text{V/K}$, one then gets $ZT > 1.6$, which is well beyond what is currently available around room temperature.

Here, we aim to directly measure the effective temperature through an electric field-driven Seebeck voltage to directly prove the existence of a locally thermal but globally nonequilibrium distribution of charge carriers in the system. To maximize the collected signal and to minimize the voltage amplitudes, we opted for structures at the nanoscale that give significant electric field strengths at low voltages. Specifically, this reduces the influence of any higher harmonic signals from, e.g., function generators or amplifiers that are unavoidably present in the driving signal and that might overwhelm the signal of interest.

As visible in Fig. 1, the experiment is set up with three electrodes: two opposing input electrodes and one perpendicular measurement electrode. In order to improve yield and identify possible trends, we manufactured 30 of such structures with varying distances per ($\sim 1.2 \times 1.2 \text{ cm}^2$) substrate. The experiment was realized on *n*-doped silicon substrates with a thermally grown 200 nm silicon dioxide cover layer. The nanostructures were manufactured by electron beam lithography in a Raith e-line pioneer system. The liftoff layer was made from PMMA 950K. The structures were developed in MIBK (4-methylpentan-2-on) and isopropanol; the liftoff was performed with NEP (n-ethyl-2-pyrrolidone). The metal layers of 5 nm of chromium and 10 nm of gold were evaporated from an efm3T electron beam evaporator. The base pressure during evaporation was $\sim 10^{-7}$ mbar. The organic films were spin coated on the lithographic structures in a glovebox at a $< 3 \text{ ppm O}_2, \text{ H}_2\text{O}$ nitrogen atmosphere. For bulk doping of regio-regular poly(3-hexylthiophene-2,5-diyl) (rr-P3HT) and 2,3,5,6-tetrafluor-7,7,8,8-tetracyanochindimethane (F_4TCNQ) were both dissolved in orthodichlorobenzene at a P3HT concentration of 20 mg/ml. All given doping ratios are molecular ratios. Depending on substrate size $\sim 60 \text{ } \mu\text{L}$ of solution was statically spin coated at 1000 rpm for 60 s and dried at 3000 rpm for 25 s. The resulting films typically had thicknesses between 60 and 100 nm. To test the behavior at high doping, sequentially doped samples were fabricated in two spincoating steps. The first consisted of a pure P3HT film as described above. In the second step 5 mg/ml F_4TCNQ were dissolved in a 4:1 mixture of tetrahydrofuran to dichloromethane of which then $\sim 80 \text{ } \mu\text{L}$ was spincoated dynamically onto the P3HT film as described by Zuo *et al.* [28]. While this is a hole transporting system, the underlying principles are equally applicable to electrons.

All experiments are performed in an N_2 -filled glovebox at room temperature (300 K).

Applying an ac voltage signal between the inputs heats the carrier distribution between them at twice the input frequency due to the direction invariance. The measurement electrode is situated outside the high field region and charge carriers in its vicinity are therefore at a lower (effective) temperature. We thus expect to measure an oscillating Seebeck voltage $\Delta V \propto S(T_{\text{eff}} - T_{\text{lat}})$ at twice the input frequency between the input ground and output [29]. Utilizing a lock-in amplifier, we can filter for these frequencies and exclude currents due to the applied potential and unintentional asymmetry in the device as these will occur at 1ω . An unwanted contribution we cannot filter via frequency is ohmic heating of the full area around the driving tips due to the input power $P = V^2/R$. As it similarly scales with the square of the applied voltage, it will also oscillate at the same frequency as the effective temperature, but it can be suppressed by using high-resistance samples. We will thus have to analyze which Seebeck voltages stem from lattice temperature differences and which are actually caused by charge carrier temperature differences.

We first discuss the raw data in order to identify the signal of interest. Representative raw data of our experiments can be found in Fig. 2, which depicts the first and second harmonic signal measured at the output electrode for a bulk doped sample. Between 10 and 1000 Hz a steep decline of the signal is visible, which we attribute to the RC cutoff of our setup. Given the high sample resistance R of $\sim 100 \text{ M}\Omega$ and the $\sim 40 \text{ pF}$ input capacitance C of our measurement device, we arrive at a cutoff frequency $f_c = 1/2\pi RC$ of $\sim 40 \text{ Hz}$, which agrees well with the observed drop in V_{out} and matches the frequency at which current and voltage measurements diverge in Fig. 2(b). This is further supported by the differing cutoffs in the sequentially

doped sample visible in Figs. S1(a) and S1(b) in the Supplemental Material (SM) with lower resistance [30].

The peak at roughly 0.1 MHz is, in contrast to the signal below 100 Hz, also visible when the structure is not coated with an organic layer; see SM Fig. S3 [30]. Therefore, we attribute it to a resonance of the structure itself and do not analyze it further. Thus, the signal of interest is situated in the low frequency regime below 100 Hz. The inset shows the scaling of first and second harmonic signals at a sufficiently low frequency of 4.75 Hz, representative of the region of interest. The sample shows a clear trend of the first harmonic signal increasing linearly. Being a first harmonic signal, this cannot be attributed to ohmic heating or effective temperature, and we attribute it to a minor asymmetry in the device, such that the symmetric drive [cf. Fig. 1(b)] does not average out to exactly zero. The relevant second harmonic signal, in contrast, increases nonlinearly as expected from the effective temperature formula Eq. (1). Being superlinear, the increase cannot be attributed to a higher harmonic in the driving signal in combination with the same geometric asymmetry as for the first harmonic signal. We can further exclude that second harmonic generation in the signal source is causing this, as the latter is measured to be several orders of magnitude smaller than the first harmonic signal, while in the device the first and second harmonics are of the same magnitude (SM Sec. 2 [30]).

As mentioned in the introduction, a possible spurious cause for a second harmonic signal is the local ohmic heating of the sample due to the applied driving voltage. As this would create a very similar, but strictly quadratic, nonlinear signal due to the dissipated power being voltage squared over resistance, we have to exclude it. To this end we first calculated the expected ohmic heating in our devices by finite element calculations, which are presented in SM Sec. 3 [30]. At low conductance, as expected for our low concentration bulk doped samples, the expected ohmic heating falls below 1 K, which is far too low to explain the measured signals. Samples with higher doping can theoretically develop significant ohmic heating and are therefore not discussed in the main text. More details can be found in SM Secs. 1 and 3 [30]. For comparison with the functional dependence expected for the effective temperature model, we have also included quadratic fits in our graphs. Where there is a difference between the two, the former gives the better description of the data.

Having identified the signal of interest as the low frequency second harmonic signal, we will now perform a quantitative analysis. We noted both a relatively large intersample variability and noiselike, i.e., stochastic, variations in individual measurements that we both attribute to the limited probing volume, making measurements sensitive to sample-to-sample variations and (field- or temperature-driven) variations in the position of individual dopants or even polymer segments, as further discussed in SM Sec. 4 [30]. We will

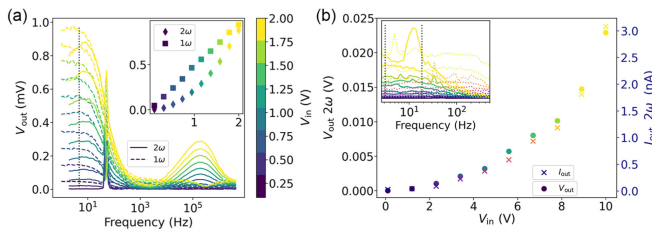


FIG. 2. (a) Raw data of first and second harmonic response measured on a P3HT film bulk doped with 0.1% molecular ratio F_4TCNQ . The input tips are separated by a 40 nm gap and the measurement tip is 60 nm from the center line. The inset shows a cut through the data at a frequency of 4.75 Hz indicated by the dotted line. The corresponding phase behavior is shown in SM Fig. S6 [30]. (b) Raw data of voltage and current output on a 1% bulk doped film averaged between 3 and 19 Hz. Here, the input and measurement distances were 15 and 33 nm. Voltage and current follow the same trend in the indicated frequency range.

focus on one representative measurement here; a more detailed discussion of remaining error sources and possible mitigation strategies is included in SM Sec. 5 [30].

Focusing on low conductivity samples brings the need to account for the high sample resistances relative to the measuring instrument. As the used lock-in amplifier has an input impedance of 10 M Ω , the actual signal is higher than measured due to the voltage division between the signal input resistance and the sample resistance and the measured voltages were corrected accordingly. More details on the used correction schemes can be found in SM Sec. 1 [30].

We are now ready to analyze the (corrected, low-frequency) data as Seebeck voltages, generated due to the temperature difference between the “hot” high-field region between the driving tips and the “cold” probe tip. For the former, we use Eq. (1), assuming $\beta = 2$ and for the latter the lattice temperature. In doing so we assumed that the applied voltage drops uniformly over the input tip separation $F \approx (V_{\text{in}}/d)$. Via numerical simulations we verified that the effect of field inhomogeneities would only lead to a correction of factor unity that is small compared to other variations between measurements and corrections (see SM Sec. 6 [30]). Hence, we arrive at the fitting expression

$$V_{\text{eff}} = S \left(\left(T_0^2 + \left(0.89 \frac{V_{\text{in}}}{d} \frac{ae}{k_b} \right)^2 \right)^{\frac{1}{2}} - T_0 \right). \quad (2)$$

For the uncorrected data presented in Fig. 3(a) this leads to a fitted localization length of 0.14 ± 1.9 Å and a fitted Seebeck coefficient of 7 ± 200 mV/K. After correcting for the resistance mismatch [Fig. 3(b)], we arrive at a localization length of 5.3 ± 1.8 Å and a Seebeck coefficient of 330 ± 190 μ V/K. As we expect localization lengths in the several Å to few nm range [37,38] and Seebeck coefficients around a few 100 s to 1000 μ V/K [28], fitted values after

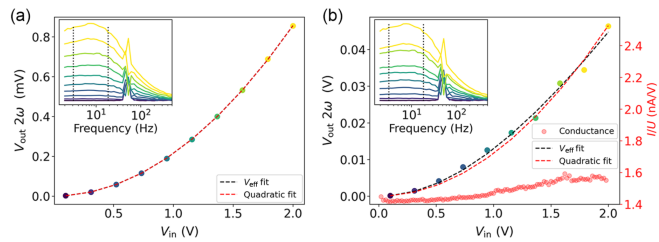


FIG. 3. (a) Cuts through the measurement fitted with Eq. (2) and a quadratic function for comparison. The P3HT films were bulk doped with 0.1% F₄TCNQ. Each point is the average between all measurements in the frequency range indicated by the black dotted lines. Plot (b) shows the same second harmonic voltage after correction for the resistance mismatch between sample and measurement device. The conductance data (red dots) utilized to calculate this correction is also displayed. The input and measurement distances were 35 and 60 nm, respectively. Corresponding current-voltage curve measured between the input tips are shown and discussed in SM Sec. 8 [30].

impedance correction are reasonable. The experimental uncertainties notwithstanding, we are convinced the modest absolute accuracy is sufficient for our main goal, that is, to prove the validity of the underlying effective temperature concept. For exact measurements of the physical parameters like conductivity or Seebeck coefficient, other, more established experiments are better suited. On uncorrected data, the effective temperature fit produces the same shape as a quadratic fit meant to describe ohmic heating. When the impedance mismatch corrections are applied, output voltage becomes more linear and is better matched by the effective temperature model.

Utilizing the fitted Seebeck coefficient, the resistance-corrected data in Fig. 3 correspond to an effective temperature about 140 K above the lattice temperature at a field of 5.7×10^7 V/m. This not only far exceeds the predicted ohmic heating of less than 1 K, it would also lead to immediate sample degradation if present in the device as an actual lattice temperature [39]. To put the number further into perspective, Nebel *et al.* have analyzed the effective temperature in doped hydrogenated silicon via its conductivity and report estimated effective hole temperatures of 180 K at a lattice temperature of 10 K and a field of $\sim 6 \times 10^7$ V/m [16]. They assumed a localization length of 5 Å for holes, well matching with our 5.25 Å, while suggesting some field dependence of the localization length might exist. Photoconductivity experiments by the same authors place the localization length at 6 Å [17]. Given that these are two different materials at different temperatures, this indicates not only a reasonable agreement, but also an (anticipated) universality across disordered systems of various types.

The measurement also excellently agrees with the effective temperatures we predicted in an earlier publication via the use of kinetic Monte Carlo simulations, as shown in Fig. 4 [23]. As the simulation data are obtained for a lattice at room temperature (300 K), a direct comparison with the experimental data is possible. Since the simulation data was created by us before the experiment, it is free from any selection bias. Moreover, and more importantly, the quantitative agreement with experiment is a very strong argument in favor of the physical reality of a field-driven enhancement of the electronic temperature in the true meaning of the word, that is, the density of occupied states forming a thermal distribution characterized by $T_{\text{eff}} \geq T_{\text{lat}}$, as (still phenomenologically) captured by Eq. (1). Since the simulation uses a framework and parameters that are generic for strongly energetically disordered semiconductors, these findings imply a general validity of the concept.

In conclusion, we utilized an ac tree-terminal experiment to test the effective temperature concept that predicts an elevated electronic temperature that is different from the lattice temperature once the energy scale associated with hopping along the electric field, eFa , becomes comparable to or larger than the thermal energy $k_B T$. As expected for periodic heating, we found a second harmonic response that

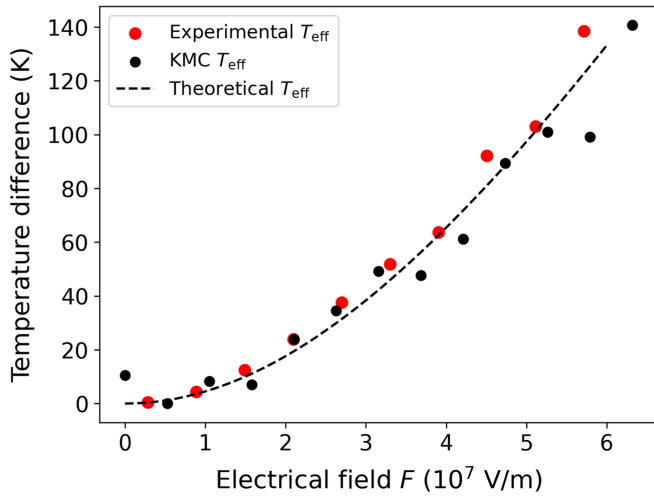


FIG. 4. Comparison of the difference between the effective electronic temperature and the lattice temperature, calculated from the experimental data of Fig. 3 (red symbols) and from kinetic Monte Carlo simulations from Ref. [23] (black symbols). The experimental data are measured (resistance-corrected) voltages divided by the fitted Seebeck coefficient. The dashed black line is the functional shape of the effective temperature formula with parameters $\alpha = 0.5$ nm, $\beta = 2$, and $\gamma = 0.89$.

corresponds to the elevated electronic temperature. By frequency-selective amplification and measuring low-doped, high-resistance films, we were able to exclude sample asymmetry and ohmic heating as spurious causes for the signal. Analyzing the measured thermovoltages with the effective temperature formalism first proposed by Marianer and Shklovskii yielded realistic values for the Seebeck coefficient and the localization length [10]. The results give a good match with previous results and an excellent match with theoretical predictions, indicating a universal applicability to strongly energetically disordered materials. An initial test of the latter can be found in SM Sec. 9 [30], suggesting that the experimental principles shown herein can indeed be transferred to other energetically disordered semiconductors, offering an experimental pathway to benchmark nonequilibrium transport models.

The experimental demonstration that the electronic temperature is largely decoupled from the lattice temperature potentially has far-reaching consequences for understanding thermalization and high-field behavior of existing device designs like solar cells and LEDs where electric fields of several V over 100 nm, i.e., in the same range as used herein, are common. While this knowledge is helpful for understanding, designing, and improving existing devices, new concepts seem possible as well. Here, we think less of conventional thermoelectric generators—even if these would greatly benefit from suppressing the losses due to lattice thermal conductivity, this would require to couple the (waste) heat source to the charge carrier distribution only, which is far from trivial. Instead, we imagine Seebeck based or enhanced devices for the

conversion of light. For instance, our results reinforce previous ideas for hot carrier solar cells that could thus be realistically based on disordered semiconductors [15,22,40]. An alternative approach might be to use plasmonic structures to couple the light field of incoming radiation into the charge carrier distribution, which is then rectified via the Seebeck effect. In all cases, the potentially large temperature differences between effective and lattice temperature, in combination with an enhanced figure of merit ZT , enable much larger power conversion efficiencies than conventional thermoelectric generators offer.

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The authors declare no conflict of interest.

Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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