Terahertz spectroscopy of charge transport in the semiconducting polymer PDPP3T

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Most high-mobility organic semiconductors are only p-conducting, which limits future device applications. Donor-acceptor polymers offer the opportunity to achieve both n-conduction and p-conduction. The charge transport in these π-conjugated polymers is not fully understood, partially because of the complex interplay between fast transport along the polymer chains, a substantially slower interchain transport, scattering, and trapping.

Terahertz electromodulation spectroscopy probes the AC conductance of mobile charge carriers and thus provides a view onto the fastest transport processes [1]. Slow processes, such as hopping motions or trapping, have only a marginal THz response. In this contribution, we present THz measurements on field-effect devices of poly-diketopyrrolopyrrole-terthiophene (PDPP3T). The structures comprise a gate contact, an insulator, a 60 nm thick layer of PDPP3T, and an MoO_x injection layer. The PDPP3T layers are fabricated by blade coating, which allows to align the nanocrystals of the polymer.

Terahertz pulses with a bandwidth of 2.5 THz are transmitted through the devices. Biasing the structures leads to the injection of charge carriers and their Drude response reduces the THz transmission. Figure 1 shows the dependence of the relative transmission $\Delta S/S$ on the bias V_g applied to the devices. Both bias polarities reduce the transmission, which proves the injection of mobile holes and electrons. Polarization dependent measurements on devices with aligned PDPP3T show that the charge transport is dominated by a fast motion along the polymer chains.

The carrier mobilities are deduced from $\mu = \frac{-\Delta S}{S} \cdot \frac{2\sqrt{\epsilon_b}}{e\, n_{2D}\, Z_0}$, where ϵ_b is the relative background permittivity and Z_0 is the impedance of free space. The charge carriers' sheet densities n_{2D} are deduced by capacitance measurements. In aligned PDPP3T, electrons and holes reach mobilities of $\mu_e = 2.2\,\mathrm{cm^2/Vs}$ and $\mu_h = 18.8\,\mathrm{cm^2/Vs}$, respectively. These values are obtained assuming that all injected charge carries are mobile and contribute to the THz signal.

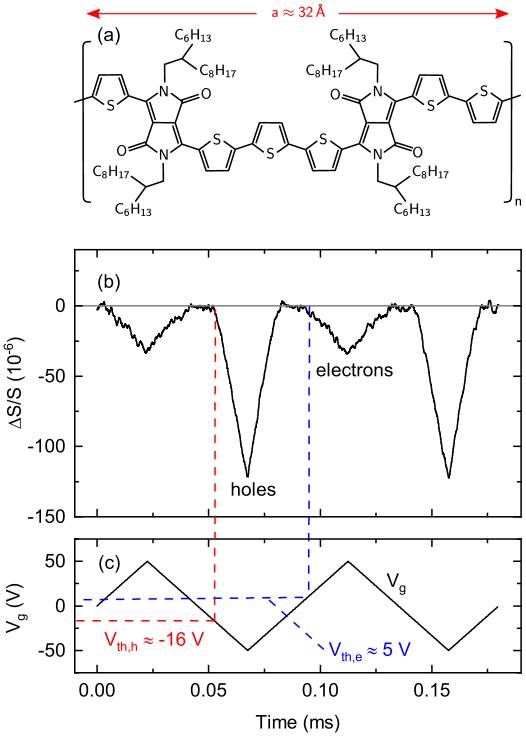

Fig. 1. (a) Unit cell of PDPP3T. (b) Relative differential transmission of THz radiation when biasing the device as shown in (c).

The carriers' mean free paths that are deduced from the mobilities are smaller than the unit cell of the polymer backbone shown in Fig. 1 (a). This, however, is not in accord with the Yoffe-Regel criterion for band transport. The contradiction is resolved, if only a fraction of the injected carriers is mobile. Consequently, carriers that propagate along the polymer chain are expected to have mobilities that by far exceed the measured values. Temperature-resolved and time-resolved measurements confirm this hypothesis [2].

References

[1] P. Riederer and R. Kersting, J. Infrared Milli. Terahertz Waves **44**, 1 (2022).

[2] P. Riederer, C. Eckel, R. T. Weitz, and R. Kersting, Appl. Phys. Lett. **123**, 182104 (2023).