## Supplementary Material to

## Effect of molecular p-doping on hole density and mobility in P3HT

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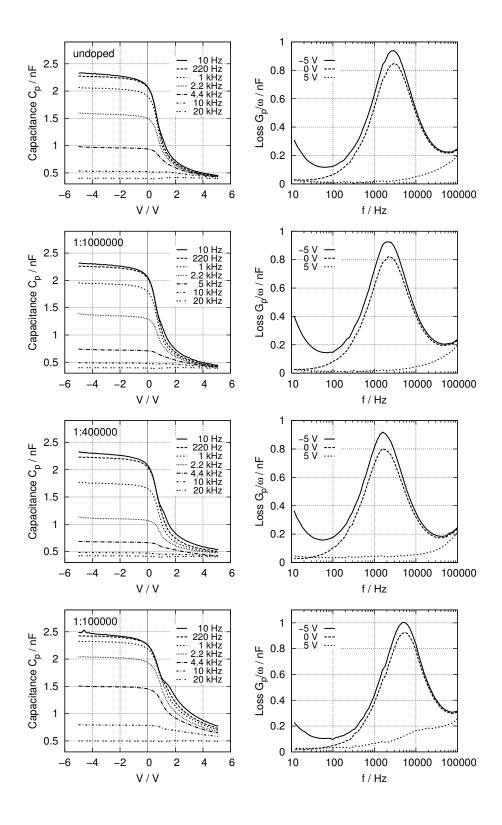
Samples for admittance spectroscopy have been prepared in a metal-insulator-semiconductor (MIS) geometry on glass substrates with patterned indium-tin-oxide (ITO). After thorough cleaning of the substrates by ultrasonication in common organic solvents, an electrically insulating layer of methyl-/phenyl-substituted polysilsesquioxane (PSQ, purchased from Gelest Inc.) was spin-cast at 1500 rpm for 30 s from a filtered 40 mg/ml solution in butanone. This layer was rendered insoluble by annealing at 350°C for 1 h. The following steps were continued in an N<sub>2</sub> glovebox under inert conditions.

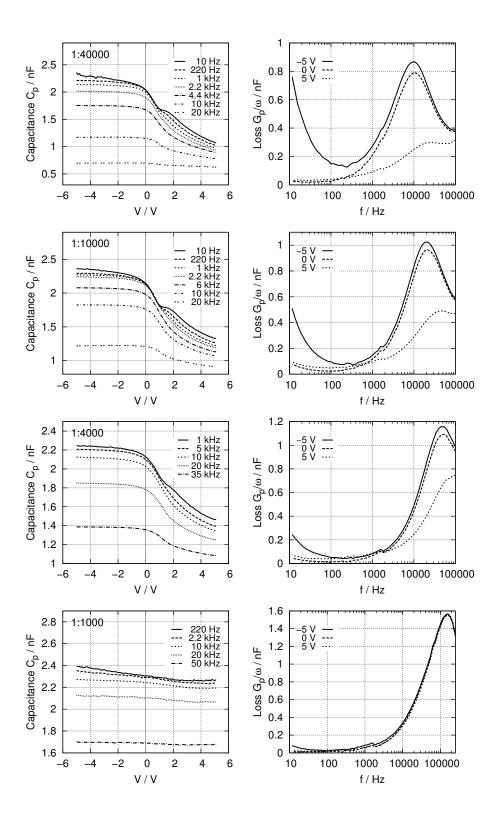
Poly(3-hexylthiophene), P3HT, and tetrafluorotetracyanoquinodimethane, F<sub>4</sub>TCNQ, were dissolved at 60 mg/ml and 0.5 mg/ml in chloroform, respectively, applying 50°C and stirring. The P3HT sample is a deuterated high-molecular weight fraction. Deuteration has been done for potential neutron scattering experiments. From our extensive previous investigations it is known that the physical properties are not altered by the deuteration and that the performance of this P3HT fraction is outstanding in field-effect devices. <sup>1–3</sup> F<sub>4</sub>TCNQ was purchased from Sigma-Aldrich. The chloroform solutions of P3HT and F<sub>4</sub>TCNQ were blended at the desired dopant concentration and processed immediately to prevent aggregation. Upon blending, the color of the solution turned immediately dark, indicative of the formation of charge transfer complexes. The semiconductor layer was then spin-cast at 1000 rpm for 30 s, yielding a visually homogeneous film. Next, electrical hole-injecting contacts were formed by evaporation of 5 nm MoO<sub>3</sub> at 2 Å/s and 100 nm Al at 3-7 Å/s. Devices were completed by encapsulation with a glass sheet and Araldite 2011 epoxy resin.

In order to determine the thicknesses of the insulator and semiconductor layers, these films were prepared on cleaned glass substrates and investigated with a DEKTAK 3 surface profilometer. Typically, the thickness of the PSQ insulator amounts to 155 nm and the thicknesses of the various doped polymer layers ranges from 350 nm to 1200 nm.

Admittance spectroscopy on MIS devices was performed using our home-built setup including an EG & G DSP 7260 lock-in amplifier and a Stanford Research SR570 low noise current amplifier. Typically, we applied an AC voltage of 20 mV to the samples.

In the following, exemplary capacitance-voltage curves and loss spectra for each of the investigated dopant concentrations are presented.





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- <sup>2</sup>A. Zen, M. Saphiannikova, D. Neher, J. Grenzer, S. Grigorian, U. Pietsch, U. Asawapirom, S. Janietz, U. Scherf, I. Lieberwirth, and G. Wegner, Macromolecules **39**, 2162 (2006).
- <sup>3</sup>P. Pingel, A. Zen, R. D. Abellon, F. C. Grozema, L. D. A. Siebbeles, and D. Neher, Adv. Funct. Mater. **20**, 2286 (2010).