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Monitoring the charge-carrier occupied density-of-states in disordered organic semiconductors under non-equilibrium conditions

Andrei Stankevych[1,2], Rishabh Saxena[2], Heinz Bässler[2], Anna Köhler[2] and Andrey Kadashchuk[1,2]

[1]*Photoactivity department, Institute of Physics of NAS of Ukraine, 03028 Kyiv, Ukraine*
[2]*Soft Matter Optoelectronics and Bavarian Polymer Institute, University of Bayreuth, 95447 Bayreuth, Germany*
Email: kadash@iop.kiev.ua and Andriy.Kadashchuk@uni-bayreuth.de

In disordered organic semiconductors, charge transport involves energetic relaxation in a broad distribution of localized states, usually assumed to be well-described by a Gaussian distribution characterized by a width σ_{DOS}. Under thermal equilibrium conditions, charges that are injected or photogenerated at a random energy site in the available density-of-states (DOS) proceed by a sequence of energetically downward or upward hops to form an occupied-density-of-states (ODOS), which is placed around an equilibrium energy ($\varepsilon_{eq} = -\sigma_{DOS}^2/k_BT$) below the centre of the DOS with a width σ_{ODOS} equal to the width of the DOS (σ_{DOS}). However, in very thin organic semiconductor layers relevant to organic optoelectronic devices or/and at low temperatures, charge-carriers are not able to reach the thermal equilibrium transport regime prior to being extracted. Therefore, the ODOS under non-equilibrium transport are expected to differ from that under equilibrium condition.

The dynamics of charge carriers in disordered organic semiconductors is inherently difficult to probe by spectroscopic methods. In the present study, we demonstrate that thermally-stimulated luminescence (TSL) technique can be used to determine the low-temperature ODOS distribution for charge-carriers. Another approach to probe charge energy relaxation are kinetic Monte-Carlo (kMC) simulations. Here we use both techniques to monitor the ODOS distribution of charges at low temperatures. We find that the charge dynamics is frustrated, yet this frustration can be overcome in TSL by using an infrared (IR) push pulse, and in kMC by a long simulation time that allows for long-range hopping transitions. Applying the IR-push TSL to pristine amorphous films of 18 commonly used low molecular weight organic light emitting diode (OLED) materials, we find that the width (σ_{ODOS}) of the ODOS universally amounts to about 2/3 of the available DOS. This implies a significant narrowing of the ODOS distribution formed at low temperatures compared to the width of the DOS. The same result is obtained in kMC simulations that consider spatial correlations between the site energies for charge carriers. Without the explicit consideration of the energetic correlations, the experimental value cannot be reproduced, which testifies to the importance of energy correlation effects for charges.

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