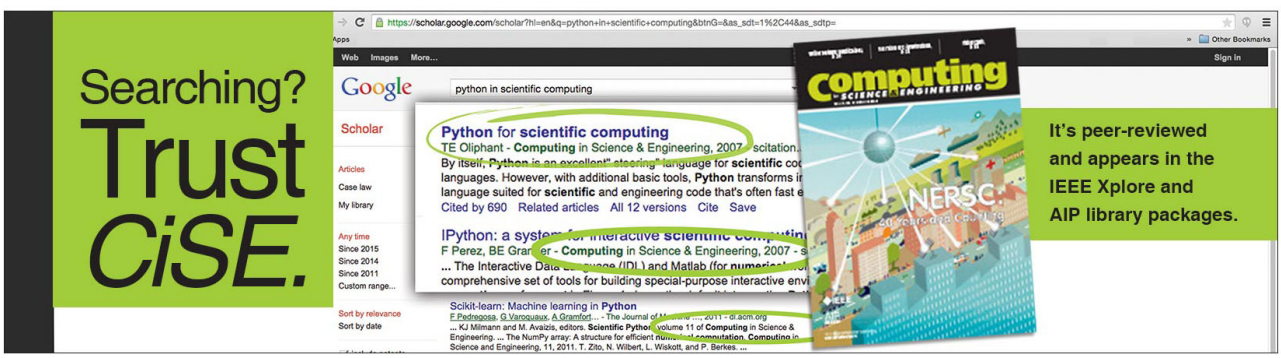
AIP | Applied Physics Letters

Effect of source-drain electric field on the Meyer–Neldel energy in organic field effect transistors

Mujeeb Ullah, A. Pivrikas, I. I. Fishchuk, A. Kadashchuk, P. Stadler, C. Simbrunner, N. S. Sariciftci, and H. Sitter

Citation: Applied Physics Letters **98**, 223301 (2011); doi: 10.1063/1.3584131
View online: http://dx.doi.org/10.1063/1.3584131
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/98/22?ver=pdfcov
Published by the AIP Publishing



Effect of source-drain electric field on the Meyer–Neldel energy in organic field effect transistors

Mujeeb Ullah,[1] A. Pivrikas,[2,a)] I. I. Fishchuk,[3,5] A. Kadashchuk,[4,5] P. Stadler,[2] C. Simbrunner,[1] N. S. Sariciftci,[2] and H. Sitter[1]
[1]*Institute of Semiconductor and Solid State Physics, Johannes Kepler University of Linz, A-4040 Linz, Austria*
[2]*Linz Institute of Organic Solar Cells, Physical Chemistry, Johannes Kepler University of Linz, A-4040 Linz, Austria*
[3]*Institute for Nuclear Research, National Academy of Sciences of Ukraine, Prospect Nauky 47, 03680 Kyiv, Ukraine*
[4]*IMEC, Kapeldreef 75, B-3001, Leuven, Belgium*
[5]*Institute of Physics, National Academy of Sciences of Ukraine, Prospect Nauky 46, 03028 Kyiv, Ukraine*

(Received 1 March 2011; accepted 2 April 2011; published online 31 May 2011)

We studied the influence of the lateral source-drain electric field on the Meyer–Neldel phenomenon observed for the charge mobility measured in C_{60}-based organic field effect transistors (OFETs). It was found that the characteristic Meyer-Neldel temperature notably shifts with applied source drain electric field. This finding is in excellent agreement with an analytic model recently extended to account also for the field dependence of the charge carrier mobility in materials with a Gaussian density-of-states distribution. As the theoretical model to predict charge carrier mobility is not limited to zero-electric field, it provides a more accurate evaluation of energetic disorder parameters from experimental data measured at arbitrary electric fields. *© 2011 American Institute of Physics*. [doi:10.1063/1.3584131]

Empirical Meyer–Neldel (MN) compensation rule[1] has been typically observed in Arrhenius plots (for the temperature dependencies) of the charge carrier mobility (μ) in organic field-effect transistors (OFETs) (Refs. 2 and 3) with changing carrier density in these devices. In context of the charge transport in OFETs, the MN rule suggests a specific empirical relation between the Arrhenius activation energy E_a and the mobility prefactor. More specifically, it implies that the Arrhenius-type [log(μ) versus $1/T$] dependencies, measured at different gate voltages and, concomitantly, at different charge carrier densities, intersect at a given finite isokinetic temperature T_{MN}. We recently studied both experimentally and theoretically the MN effect in C_{60} OFETs and have shown that the characteristic parameter called MN energy "$E_{MN}=kT_{MN}$" is determined by the width of the density-of-state (DOS) distribution, σ, in the conductive channel of an organic semiconducting film.[4–6] Measurements of the MN effect in C_{60} films grown at different conditions have revealed a significant shift in the MN energy (and consequently change in the energetic disorder parameter σ) with changing film morphology.[5] Thus it was proposed[5] that "E_{MN}" can be used as an important material characterizing parameter for active organic semiconductor layers in OFETs independent of the device geometry.

The initial theoretical consideration of the MN effect[4] in organic materials was based on a Gaussian disorder model with accounting for the carrier concentration dependence of the OFET mobility, however, it was limited to zero-electric field implying thus it is justified just for a very low lateral field. Recently Fishchuk *et al.*[7] extended the above theoretical model to consider the OFET mobility at arbitrary electric fields, viz., lateral field caused by source-drain voltage. In the present letter, we report on experimental and theoretical investigation of the influence of the lateral electric field on the MN energy in OFETs. The shift in the MN temperature in an OFET upon applied lateral electric field was found and described by the extended analytic model of Fishchuk *et al.*[7] that accounts for the field dependence of the OFET mobility. We show that the observed electric field dependence of the MN energy is a consequence of the spatial energy correlations in the organic semiconductor film which features a Poole–Frenkel type behavior ($\ln \mu \propto F^{1/2}$) for OFET mobility upon applied source-drain field. Since the present model is not limited to zero-field, it allows more accurate evaluation of energetic disorder parameters from experimental data measured at a given electric field.

We investigated in C_{60} based OFET structure the temperature dependence of charge carrier mobility which was evaluated from the transfer characteristics in the linear regime ($V_g \gg V_d$, where V_g and V_d are the gate and drain source voltages, respectively) in order to ensure a homogeneous charge distribution in the conductive channel. In order to maintain the linear regime conditions we used low source drain voltages. These OFET devices were fabricated using divinyltetramethyldisiloxane-bis(benzocyclobutane) as gate dielectric on ITO/glass substrates and C_{60} thin films as organic semiconductor as described previously.[2]

The observation of different Arrhenius activation energies of electron mobility for OFET at different carrier concentrations leads to the phenomenological MN rule. Figure 1(a) shows the MN type behavior of charge carrier mobility at a source-drain voltage of 2 V and Fig. 1(b) shows the MN behavior at a source-drain voltage of 10 V. A shift in the MN temperature from 409 to 372 K is clearly visible in both Arrhenius plots Figs. 1(a) and 1(b).

As shown in Fig. 2 the Arrhenius activation energy of charge carrier mobility decreases with increasing V_g, which

a)Electronic mail: almantas.pivrikas@jku.at.

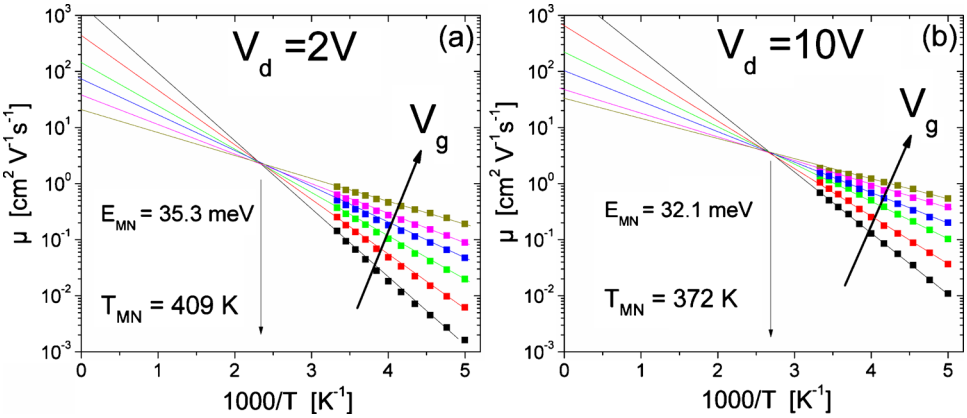


FIG. 1. (Color online) Meyer Neldel rule behavior of charge-carrier mobility measured at different gate voltages for two applied source –drain voltages (a) 2V (b) 10 V. Range for V_g is from 25 to 60 V.

can be explained by the fact of filling up the DOS with increasing charge carrier concentration and consequently shifting the Fermi level closer to the effective transport energy level.[8,9] This decreases the activation energy, charge carriers has to overcome in order to be mobile.

The activation energy decreases with increasing electric field in the film, which can be explained by the fact, that the electric field lowers the average barrier height for energetic uphill jumps in the field direction.[10] Figure 2 shows a Poole–Frenkel type behavior of the activation energy which decreases linearly with square-root of the source-drain voltage.

The MN energies measured in C_{60} OFETs at different source-drain voltages are plotted in Fig. 3 (symbols). The E_{MN} was found to shift from 35 to 32 meV with increasing lateral electric field in the device.

Recently, we extended our analytic model, which was originally suggested for a zero-field limit, to consider the temperature dependent OFET mobility also at *arbitrary electric fields*.[7] The model is based on effective medium formalism and takes also into account spatial energy correlation effects using the results of recent computer simulations[11] of charge-carrier transport in energy correlated system at large carrier concentrations (see for calculation details in Ref. 7). Energy correlations in organic disordered solids imply slowly varying static spatial fluctuation in the potential energy landscape and can arise due to charge-dipole[12] or charge-quadrupole interactions or fluctuations (inhomogenity) in electronic polarization energy, resulting from molecular density fluctuations in an organic material due to microscopic regions that are under compression or dilation.[13]

In particular, the analytic model[7] accounts for a reduced *local* variance in the energy distribution of nearby hopping sites due to the correlation-induced smoothing of the energy landscape in a disordered organic solid and, what was found to be of special relevance for the description of the presented experimental results, is that it accounts for a decrease in the slope of the Pool–Frenkel-type field dependence with increasing carrier density.[11] The latter was found to be an immediate reason giving rise to the lateral field (F) dependence of the MN energy. This dependence can be parametrized in the following form[7] for $a/b=5$

$$\frac{E_{\mathrm{MN}}}{\sigma} = 0.5 + 0.029\frac{eaF}{\sigma} - 0.039\left(\frac{eaF}{\sigma}\right)^2. \tag{1}$$

where e is the elementary charge, a is an average intermolecular distance assumed for C_{60} films to be similar to C_{60} crystals, viz., $a=1.4$ nm,[14] and b is the carrier localization radius. The width of the DOS σ can be determined as

$$\sigma = E_{\mathrm{MN}} A\left[1 + \sqrt{1 + \left(\frac{eaF}{E_{\mathrm{MN}}}\right)^2 \frac{0.078}{A^2}}\right], \quad A = 1 - 0.029\frac{eaF}{E_{\mathrm{MN}}}. \tag{2}$$

Equations (1) and (2) are valid for $eaF/\sigma > 0.25$, i.e., when the Poole–Frenkel-type field dependence ($\ln \mu \propto F^{1/2}$) of the

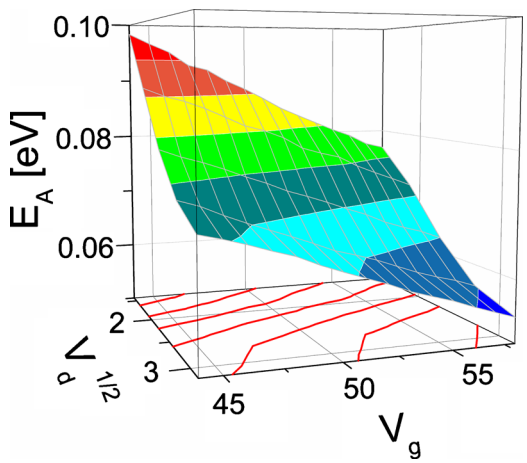


FIG. 2. (Color online) Arrhenius activation energy measured as a function of gate voltage V_g at different applied source-drain voltages V_d.

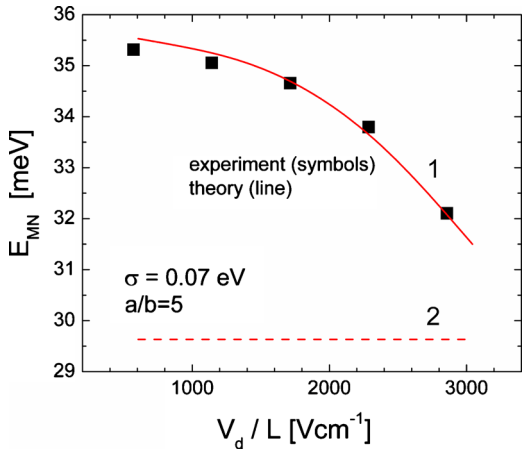


FIG. 3. (Color online) Meyer—Neldel energy as a function of the lateral electric field measured in C_{60}-based OFET (symbols) and the theoretical fit based on our extended model as described by Eq. (1) and (2). Dashed curve 2 was calculated by neglecting any energy correlation effects.

OFET mobility is obeyed. At lower fields, the charge mobility tends to saturate upon approaching zero-field and thus deviates from the Poole–Frenkel law.[7,11] Consequently, the actual zero-field mobility differs from that obtained by extrapolations of $\ln \mu \propto F^{1/2}$ plots to $F \to 0$. It should be noted that present extended theoretical model yields $E_{MN}/\sigma \cong 0.33$ for zero-field mobility. Previous theoretical treatment limited to zero-field case,[4] which disregarded the energy correlations and percolation effects, yielded a somewhat different ratio $E_{MN}/\sigma \cong 0.40$.

Thus, the experimentally determined E_{MN} at a finite electric field can directly be used for evaluation of the energy disorder parameter σ by Eq. (2) without the necessity of an extrapolation of the experimental data to zero-electric field.

It should be noted, that for the quantitative description of the lateral field dependence of the OFET mobility one has to involve a concept of a strong inhomogenity of the lateral electric field inside the accumulation layer formed in an OFET caused by an inhomogeneous morphology of the semiconductor, which results in strong local fields confined to specific places having the largest energetic barrier heights and, hence, controlling the overall (effective) hopping charge mobility through the OFET channel, as suggested elsewhere.[7] Thus, it is actually the effective local field that should be used in Eq. (2) instead of the average applied electric field. The ratio q between the local electric field strength and that averaged over the transistor channel V_d/L can be readily determined from the field dependence of E_{MN}. In fact, a set of just two E_{MN} values measured at two different electric fields inserted into Eq. (2) enables calculating σ and q parameters.

Solid curve 1 in Fig. 3 shows the calculated lateral electric field dependence of E_{MN} using Eqs. (1) and (2) at $q=256$ and $a=1.4$ nm, and it demonstrates a remarkably good fitting of the experimental data by the present analytic model for $F=V_d/L>500$ V cm^{-1}, when $\ln \mu \propto F^{1/2}$ dependence for the OFET mobility takes place. The calculated field dependence of E_{MN} for the system devoid of any spatial energy correlations is given by dashed curve 2 in Fig. 3. Thus, the experimentally observed decrease in the MN energy with increasing electric field results from presence of spatial energy correlations.

In conclusion, the MN behavior for the temperature dependent OFET mobility has been studied in C_{60} films at different applied lateral electric fields. The characteristic MN energy E_{MN} is found to shift from 35 meV $\to$ 32 meV by increasing the applied source drain electric field. The experimental results are in excellent agreement with the predictions of the recent analytic model[7] which was extended to account for the dependence of the OFET mobility upon the applied lateral electric field in organic semiconductors with a Gaussian DOS distribution. The present model is superior to the previously suggested one as it allows more accurate evaluation of important material parameters from experimental data measured at any electric field and does not require an extrapolation of experimental data to the zero-electric fields.

The research was implemented within the bilateral ÖAD Project No. UA-10/2011 and supported by the Ministry of Education and Science of Ukraine (Project No. M/125-2009), the Austrian Science Foundation (NFN Project Nos. S9706 and S9711), and the Science&Technology Center in Ukraine under Contract No. 5258.

[1]W. Meyer and H. Neldel, Z. Tech. Phys. (Leipzig) **18**, 588 (1937).
[2]M. Ullah, T. B. Singh, H. Sitter, and N. S. Sariciftci, Appl. Phys. A: Mater. Sci. Process. **97**, 521 (2009).
[3]E. J. Meijer, M. Matters, P. T. Herwig, D. M. de Leeuw, and T. M. Klapwijk, Appl. Phys. Lett. **76**, 3433 (2000).
[4]I. I. Fishchuk, A. Kadashchuk, J. Genoe, M. Ullah, H. Sitter, T. B. Singh, N. S. Sariciftci, and H. Bässler, Phys. Rev. B **81**, 045202 (2010).
[5]M. Ullah, I. I. Fishchuk, A. Kadashchuk, P. Stadler, A. Pivrikas, C. Simbrunner, V. N. Poroshin, N. S. Sariciftci, and H. Sitter, Appl. Phys. Lett. **96**, 213306 (2010).
[6]A. Pivrikas, M. Ullah, T. B. Singh, C. Simbrunner, G. J. Matt, H. Sitter, and N. S. Sariciftci, Org. Electron. **12**, 161 (2011).
[7]I. I. Fishchuk, A. Kadashchuk, M. Ullah, H. Sitter, S. Sariciftci, and H. Bässler, "Electric field dependence of charge-carrier hopping transport within the random energy landscape in an organic field effect transistor," Phys. Rev. B (to be published).
[8]W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. de Leeuw, and M. A. J. Michels, Phys. Rev. Lett. **94**, 206601 (2005).
[9]I. I. Fishchuk, V. I. Arkhipov, A. Kadashchuk, P. Heremans, and H. Bässler, Phys. Rev. B **76**, 045210 (2007).
[10]H. Bässler, Phys. Status Solidi B **175**, 15 (1993).
[11]M. Bouhassoune, S. L. M. van Mensfoort, P. A. Bobbert, and R. Coehoorn, Org. Electron. **10**, 437 (2009).
[12]S. V. Novikov, D. H. Dunlap, V. M. Kenkre, P. E. Parris, and A. V. Vannikov, Phys. Rev. Lett. **81**, 4472 (1998).
[13]S. V. Rakhmanova and E. M. Conwell, Appl. Phys. Lett. **76**, 3822 (2000).
[14]H.-B. Bürgi, E. Blanc, D. Schwarzenbach, S. Liu, Y.-J. Lu, M. M. Kappes, and J. A. Ibers, Angew. Chem. Int. Ed. Engl. **31**, 640 (1992).