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## Electric field dependent activation energy of electron transport in fullerene diodes and field effect transistors: Gill's law

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The electric field and temperature dependence of the electron mobility is studied comparatively in the bulk of fullerene (C<sub>60</sub>) diodes and at the interface with dielectric of organic field effect transistors (OFETs). Electron mobility values follow a Poole–Frenkel-type electric field dependence in both types of devices. The activation energy for electron transport is electric field dependent and follows the square root law of field in both devices as predicted by Gill's law. The same Gill's energy  $E_{\text{Gill}}=34$  meV is measured in diodes and OFETs, which corresponds well to Meyer–Neldel energy ( $E_{\text{MN}}=35$  meV). It is shown that both the electric field and charge carrier concentration must be accounted for the description of disordered charge transport. © 2011 American Institute of Physics. [doi:10.1063/1.3557503]

Charge carrier transport, even though being a material property, can strongly differ in various devices.<sup>1,2</sup> Therefore, comparative studies in various device configurations are required in order to fully understand charge transport phenomena and to develop general theoretical models.<sup>3</sup> Carrier concentration, electric field, and temperature dependent carrier mobility are a signature of disordered transport. Bässler's model is often used to describe the electric field and temperature dependent mobility; however, carrier concentration is assumed to be constant.<sup>4</sup> Carrier density dependent mobility was analyzed in terms of Meyer–Neldel rule (MNR) in the past in fullerene devices, and it was shown that MNR energy can be used to characterize the degree of disorder in the C<sub>60</sub> films.<sup>5–10</sup> In general, Meyer–Neldel rule describes temperature activated process, where Arrhenius activation law is observed and the activation energy depends on a certain parameter, such as carrier density, mobility, electric field, etc.<sup>11</sup> In the past it was shown that charge carrier density has a significant effect on the activation energy which results in MNR in fullerene.<sup>5</sup> In addition to carrier concentration dependent charge transport, Poole–Frenkel-type electric field dependent mobility ( $\ln \mu = SE^{1/2}$ , where  $S$  is the proportionality factor) is usually observed in disordered semiconductors.<sup>12,13</sup> The effect arises from a reduction in the thermal activation energy necessary to activate carriers due to lowering the Coulomb barrier by the external electric field. A deviation of both the magnitude of  $S$  and its temperature dependence from the prediction of Poole–Frenkel theory even including a negative sign of  $S$  is generally seen in organic semiconductors.<sup>14</sup> The temperature dependence of the slope  $S$  has been first observed in trinitrofluorenone and poly-n-vinylcarbazole by Gill and later in other materials.<sup>15–18</sup> The empirical relation for electric field and temperature dependence charge carrier mobility has been proposed by Gill:<sup>15</sup>

$$\mu = \mu_0 \exp[-(\beta\sqrt{E})/kT_{\text{eff}}], \quad \frac{1}{T_{\text{eff}}} = \frac{1}{T} - \frac{1}{T_{\text{Gill}}}, \quad (1)$$

where  $\mu_0$  is the mobility prefactor,  $\beta$  is the Poole–Frenkel parameter,  $E$  is an applied electric field,  $k$  is the Boltzmann constant,  $T$  is the temperature, and  $T_{\text{Gill}}$  is the temperature at which the mobility is independent of the electric field.

In this work, previously studied carrier density dependent charge transport, showing MNR in fullerene devices, is extended to account for electric field and temperature dependent electron mobility obeying Gill's law.<sup>5</sup> Two different devices, diodes and organic field effect transistor (OFET), were studied where charge carrier mobility in diodes was measured using where charge extraction with linearly increasing voltage (CELIV). Due to the significant amount of equilibrium charge carriers, classical time-of-flight technique could not be applied in the studied films.<sup>19</sup>

In order to simplify the carrier mobility calculations from CELIV, the experimental conditions were chosen to ensure photogeneration at the surface by applying lowest possible laser intensity.<sup>20</sup> Thick fullerene films ( $\approx 1-10$   $\mu\text{m}$ ) were grown on indium tin oxide (ITO)/glass substrates using physical vapor deposition. Semitransparent 30 nm thick aluminum electrode was vacuum evaporated on top of the fullerene films. Diodes could only be measured down to 200 K due to the peel-off of thick fullerene films at lower temperatures.

In OFET studies, charge carrier mobility was measured from transfer characteristics in the linear regime ( $V_G \gg V_{\text{DS}}$ , where  $V_G$  and  $V_{\text{DS}}$  are the gate and drain-source voltages, respectively) in order to ensure a homogeneous charge distribution in the channel. OFET devices were fabricated using divinyltetramethyldisiloxane-bis (benzocyclobutane) (BCB) as gate dielectric on ITO/glass substrates and fullerene thin films as organic semiconductor.<sup>21</sup>

In both CELIV and OFET experiments, the charge carrier mobility and its temperature dependencies were measured in an Oxford nitrogen cryostat under vacuum below  $10^{-6}$  mbar. Fullerene devices were never exposed to air. The

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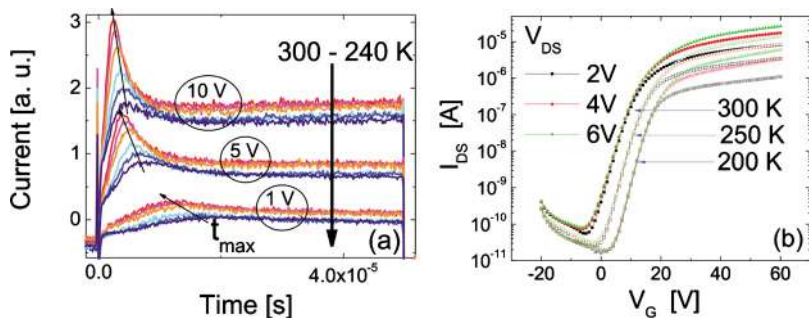


FIG. 1. (Color online) (a) CELIV photocurrent transients (delay time between the laser pulse and triangle voltage is  $0.1 \mu\text{s}$ ) shown for different temperatures as well as for various maximum voltages of triangle voltage pulse. Thin arrows mark the extraction maximum  $t_{\text{max}}$  from which electric field dependent mobility is calculated. The applied temperature range is shown by thick arrow. (b) OFET transfer characteristics for different source-drain voltages and temperatures.

electron mobility in the studied films has turned out to be much higher than hole mobility; therefore, in this study we have measured the properties of electron transport.<sup>5</sup> The following relation was used to calculate the carrier mobility in thick films:  $\mu = Kd^2 / (At_{\text{max}})$ , where  $d$  is the film thickness,  $K=2$  for surface photogeneration ( $\alpha d \gg 1$ , where  $\alpha$  is the light absorption coefficient from the Beer–Lambert law),  $t_{\text{max}}$  is the time when the extracted current has reached its maximum, and  $A$  is the rate of change of the applied voltage.

Figure 1(a) shows CELIV transients recorded in the fullerene diodes at various applied electric fields and different temperatures. From these transients, the electric field dependent electron mobility is calculated. It is in the nature of the CELIV method that the field is linearly increasing during the experiment, and thus it is a not well defined constant. However, since the strongest extraction happens at  $t_{\text{max}}$ , the electric field is estimated at the extraction maximum ( $At_{\text{max}}/d$ ). A Poole–Frenkel-type dependence of the mobility is seen for all temperatures and electric fields in both devices. The extraction maximum shows a stronger temperature dependence at lower applied voltages [as shown by the arrows marking  $t_{\text{max}}$  in Fig. 1(a)], which directly indicates stronger temperature dependence of the mobility at lower fields.

Figure 1(b) shows OFET transfer characteristics as a function of the applied source-drain bias at different temperatures. The electric field dependent carrier mobility was evaluated in the linear regime. An on-off ratio higher than five orders of magnitude can be seen as it was also observed before.

The electron mobility for both diode and OFET is shown in Fig. 2(a) as a function of the applied electric field at different temperatures. More than one order of magnitude higher electron mobility values are observed in OFETs. It is important to note that the charge carrier concentration differs by more than one order of magnitude in both experiments due to the two dimensional nature of the carrier concentration distribution in the channel of the transistor. In CELIV the concentration range is between  $2 \times 10^{15}$  and  $8 \times 10^{15} \text{ cm}^{-3}$ , whereas in OFET the range is  $2 \times 10^{17}$ – $5 \times 10^{17} \text{ cm}^{-3}$ . It was tried to experimentally limit the large concentration difference by lowering the gate voltage but still working in the linear regime of OFETs. In CELIV experiments, the carrier concentration is limited by the charge stored on the electrodes  $CV$  (capacitance times voltage) due to Langevin type of bimolecular carrier recombination at high concentrations in fullerene.

As it can be seen from Fig. 2(a) the electric field dependence increases at lower temperatures. The extrapolated mobility data points converge to an electric field, where the

carrier mobility becomes temperature independent,  $F_{\text{CELIV}} = 9.2 \text{ kV/cm}$  and  $F_{\text{OFET}} = 9.4 \text{ kV/cm}$ .

In order to demonstrate Gill’s law, the carrier mobility data from Fig. 2(a) are graphically replotted in Arrhenius plot as shown in Fig. 2(b). Important observations are that (a) the data follow an Arrhenius-type dependence (in contrast to polymers, where  $1/T^2$  temperature dependence is usually seen and explained by the Bässler model), (b) the carrier mobility is an activated process, (c) the activation energy is field dependent (decreasing with increasing field), and (d) the extrapolated data intersect at  $T_{\text{Gill}}$ .

The isokinetic point ( $T_{\text{Gill}}$ ) shows the energy  $E_{\text{Gill}} = kT_{\text{Gill}}$  required for electric field independent carrier mobility. The corresponding Gill’s energies are  $E_{\text{CELIV}} = 34 \text{ meV}$  and  $E_{\text{OFET}} = 34 \text{ meV}$ . The experimentally observed  $E_{\text{Gill}}$  value is similar to the Meyer–Neldel energy  $E_{\text{MN}} = 35 \text{ meV}$  observed in the past in fullerene films.<sup>5</sup>

From the Arrhenius plot the activation energy at different electric fields was evaluated and is shown in Fig. 3. In

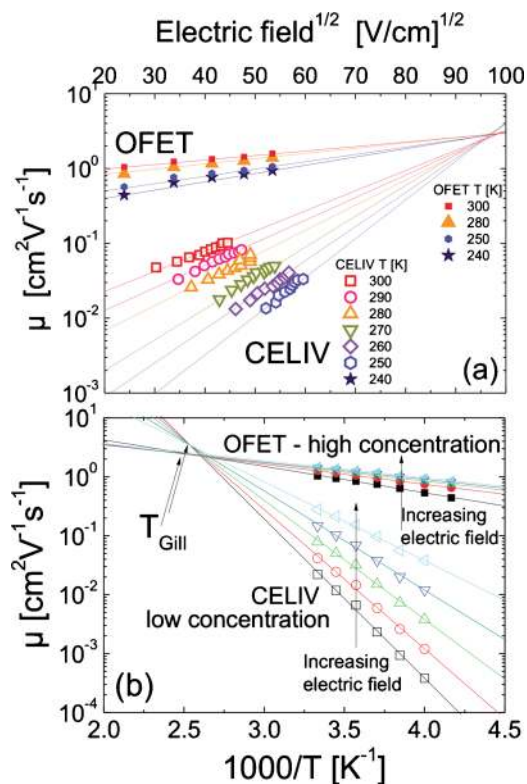


FIG. 2. (Color online) (a) Poole–Frenkel plot of electron mobility at various temperatures measured from CELIV and OFET. (b) Arrhenius plots of electron mobility at various electric fields. Gill’s temperature  $T_{\text{Gill}}$  is shown for both types of devices.



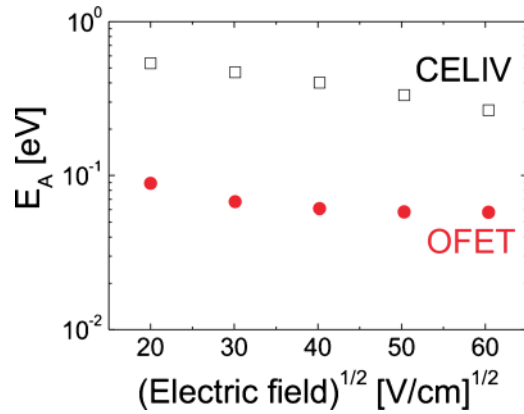


FIG. 3. (Color online) Activation energy plotted in the Poole–Frenkel law as a function of square root of electric field. More than order of magnitude larger charge carrier concentration is present in OFETs.

agreement with studies in the past, the activation energy follows a square root dependence on electric field in diodes within the range of experimental error.<sup>15</sup>

As described above, from disordered transport formalism in comparison to diode structures, a much lower activation energy is required in OFETs due to a much larger charge carrier concentration present in the channel of field effect transistor. Detailed theoretical models accounting for electric field, carrier concentration, and temperature dependent mobility are required in order to explain the observed experimental results.

In conclusion, it is shown that the electron mobility follows a Poole–Frenkel-type electric field dependence in fullerene diodes and field effect transistors. A comparison of carrier mobility in both types of devices reveals that electron mobility is determined by both carrier density and electric field as seen from much higher electron mobilities in the channel of OFETs. Gill's law is observed in both types of devices showing that activation energy for electron transport is larger at lower applied fields. Gill's energy value in both devices,  $E_{\text{Gill}} \approx 34$  meV, is similar to Meyer–Neldel energy  $E_{\text{MN}} = 35$  meV observed in the past. Larger activation energy for electron transport is required in diodes at lower carrier

concentrations in comparison to OFETs, which concludes that electric field and carrier concentration shall be accounted for when describing charge transport in disordered semiconductors.

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