Switching in C_{60}-fullerene based field effect transistors

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We are reporting on the electrical properties of a bottom gate C_{60}-fullerene based *n*-channel organic field effect transistor. The C_{60} thin film was epitaxially grown using hot wall epitaxy on top of an organic dielectric divinyltetramethyldisiloxane-bis(benzocyclobutene). The device performance depends on the growth parameters during the C_{60} film growth. Optimization of the growth parameters leads to a C_{60} film of a low total number of traps, and the drain-source current is increased by two orders in magnitude. We propose that the high current-densities are caused by space charge limited currents beside the gate induced space charge. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2216869]

Fullerenes exhibit a very special case of organic compounds which may possess semiconducting, metallic, and even superconducting properties.[1–3] In this letter, we present results on organic field effect transistors (OFETs) based on C_{60} grown on top of organic dielectrics using hot wall epitaxy (HWE). The HWE can be maintained to operate close to the thermodynamic equilibrium. Consequently the molecule can find the most suitable arrangement before being condensed, resulting in highly ordered structures of the deposited layer.[4,5] Electron mobilities up to 6000 $cm^2/V\,s$ have been measured in barium doped crystalline C_{60} films grown by the HWE technique.[6] The controlled growth conditions ensures the preparation of a defined C_{60} thin film morphology. Thus the electrical properties of the C_{60} thin film are alterable in a reproducible way. Optimization of the growth-parameters enhances the drain-source current to values up to 98.9 mA at a drain-source voltage of 100 V and at a gate-source voltage of 75 V.

The geometric structure of the OFET is shown in Fig. 1. On top of a pattered indium-tin-oxide glass (ITO-glass) substrate, divinyltetramethyldisiloxane-bis(benzocyclobutene) (BCB) from Dow Chemicals as organic dielectric was spin coated under ambient conditions. The resulting BCB film thickness was ≈ 2 μm with a surface roughness below 5 nm. After spin coating, the BCB was thermally cross-linked for 30 min at 250 °C in Ar atmosphere. On top of the BCB a 300 nm thick C_{60} was grown using HWE where once the BCB was *in situ* preheated prior to film growth at 250 °C for 20 min and in the other case not. For all samples after thermalization the substrate was kept at a constant of 130 °C during the C_{60} film growth. Finally the LiF/Al (0.6/60 nm) drain-source electrodes were prepared via evaporation through a shadow mask under dynamic vacuum of $\approx 10^{-6}$ mbar.

The output characteristics in accumulation mode of the drain-source currents versus the drain-source voltages $I_{DS}(V_{DS})$ are substantially different when the C_{60} film was grown on a preheated or on a nonpreheated dielectric. In Fig. 2 where C_{60} was grown on a nonpreheated dielectric the $I_{DS}(V_{DS})$ characteristics features a well defined pinch off voltage ($V_{DS} \approx V_{GS}$). This is followed by a high differential resistance $r = \partial V_{DS}/\partial I_{DS}$ in the saturation regime ($V_{DS} > V_{GS}$) of the output characteristics. The well defined pinch off voltage allows the use of the conventional metal-oxide-semiconductor field-effect transitor (MOSFET) relationship between the mobility and the channel's transconductance in the saturation regime and is given by[7]

$$\mu_{eff} = \frac{L}{V_{GS} W C_i} \frac{\partial I_{DS}}{\partial V_{GS}}, \qquad (1)$$

where μ_{eff} is the field effect mobility of the free charge carriers n_f, C_i is the capacitance of the gate per unit area, and L and W are the channel length and width, respectively. From the parameter $W = 1.4$ mm, $L = 35$ μm, and $C_i = 1.2$ nF/cm^2 a field effect mobility of $\mu_{eff} \approx 0.6$ $cm^2/V\,s$ is calculated.[8] In contrast, the output characteristics for the OFET with C_{60} grown on a preheated dielectric shows an increased total drain-source current and a reduced differential resistance in the saturation regime (see inset of Fig. 3). The differential resistance r in the saturation regime up to a gate-source voltage of $V_{GS} = 20$ V does not change significantly. Further increase of the gate-source voltage decreased r resulting in a vanishing saturation of the $I_{DS}(V_{DS})$ current. At a gate-source voltage of $V_{GS} = 75$ V and above a drain-source voltage of $V_{DS} = 98$ V the drain-source current increased two orders in magnitude and reached values as high as $I_{DS} = 98.9$ mA at $V_{DS} = 100$ V. This dramatic increase of I_{DS} was routinely observed in a series of samples and appears to be reversible and

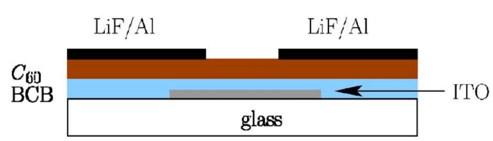

FIG. 1. (Color online) Geometric structure of the C_{60} organic field effect transistor.

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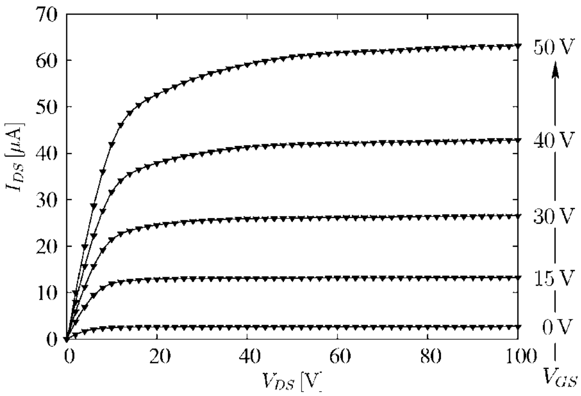

FIG. 2. $I_{DS}(V_{DS})$ characteristics of a C_{60} based OFET in accumulation mode. The C_{60} was grown using HWE on a nonpreheated dielectric.

not caused by an irreversible "breakdown" of the OFET.

Without any assumptions of a particular transport model Koehler and Biaggio[9] derived a general equation for the saturated drain-source current I_{SAT} vs V_{GS} and is given by

$$I_{SAT} = \frac{W}{2L} C_i V_{GS}^2 \mu_{eff} \left(1 + \frac{2C_i}{3Q_t} V_{GS}\right), \quad \mu_{eff} = \mu \frac{\alpha}{Q_t}, \qquad (2)$$

where $\alpha = \beta / \gamma$. γ and β are related to the emission and capture rates in/out of the traps $Q_t = eN_tD$, N_t is the average density of trap centers per volume, D is the C_{60} film thickness, and μ is the microscopic mobility. The other variables have the same meaning as in Eq. (1). By neglecting the second summand in the brackets of Eq. (2), Eq. (2) reduces to Eq. (1). Similar equations have been derived by Horowitz and Delannoy[10] by assuming a transport band and a single trap level in the band gap. The effective mobility therein is $\mu_{eff} = \mu\theta$, where θ is the fraction of the total gate induced space charge which is free and μ is the microscopic mobility. Any field dependency of μ is not addressed in those models but can be important when conjugated polymers are used.[11,18]

The difference of the output characteristics in Figs. 2 and 3 are interpreted due to the lower average density of traps for the C_{60} film grown on the preheated dielectric. Figure 4 shows two atomic force microscopy (AFM) topographic images of C_{60} films grown on a preheated as on a nonpreheated dielectric, respectively. Both C_{60} films appear to be polycrystalline. The variation of the grain size is decreased and the total grain size is increased for C_{60} grown on the preheated dielectric [Fig. 4(b)] compared with the film grown on the nonpreheated dielectric [Fig. 4(a)]. The lower overall entropy of the C_{60} film grown on the preheated dielectric caused a decrease in the total number of traps. Consequently [after Eq. (2)] the effective mobility μ_{eff} is increased. It is noted that Eq. (2) has been derived assuming a higher average density of traps N_t compared to the gate induced trapped charge-carrier concentration N_g ($N_t \gg N_g$). This assumption is not generally fulfilled at high gate-source voltages and for a high quality organic semiconductors. Furthermore, Eq. (2) does not describe the dependency of the saturated current on the drain-source voltage [$I_{DS}(V_{DS})$, $V_{DS} > V_{GS}$].

Generally the saturation of the $I_{DS}(V_{DS})$ current in a OFET is caused by the highly resistive part of the channel close to the drain contact. In this part of the channel, the gate potential is higher than the potential of the free charge carriers. This causes the free charge carriers n_f to be pushed

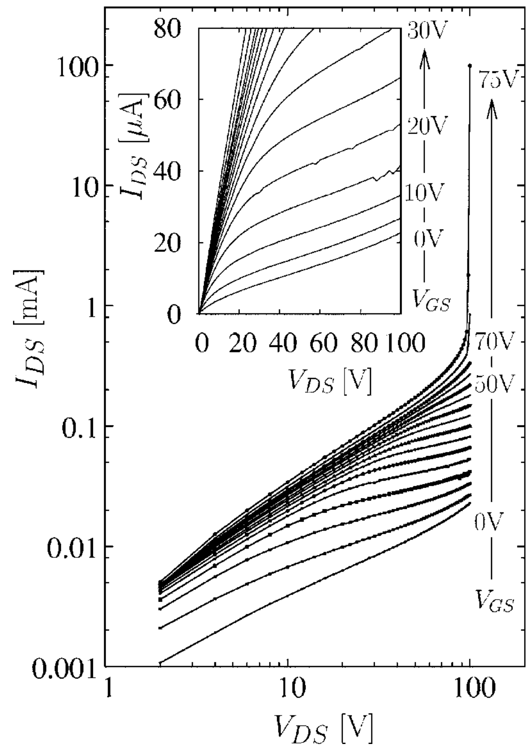

FIG. 3. Double logarithmic presentation of a $I_{DS}(V_{DS})$ OFET characteristics in accumulation mode. The C_{60} was grown using HWE on a *in situ* preheated dielectric. At $V_{GS} > 50$ V the $I_{DS}(V_{GS})$ is not saturating and reaches values of $I_{DS} = 98.9$ mA at $V_{GS} = 100$ V and $V_{GS} = 75$ V.

away from the interface between the gate dielectric and the C_{60} film (depleting the channel) and n_f reaches values of the intrinsic material. The low n_f concentration close to the drain contact results in a high electric field E parallel as well as perpendicular to the C_{60} film which ensures the constancy of the total current ($\nabla I_{DS} = 0$) within the channel. Increasing the gate-source voltage V_{GS} (relative to the drain-source voltage V_{DS}) decreases the size of the depleted region and the depleted region vanishes when $V_{GS} > V_{GS}$. As a consequence the differential resistance of the saturated drain-source current is directly related to the conductivity in the depleted part of the channel. Deviations of the linearity of the saturated current versus the drain-source voltage are predominantly caused by the nature of the transport in the depleted region of the channel.

As noted in Ref. 9 space charge limited currents (SCLC) caused by the drain-source current beside the space charge induced by the gate can significantly influence the OFET characteristics. As widely discussed in the literature[12–17]

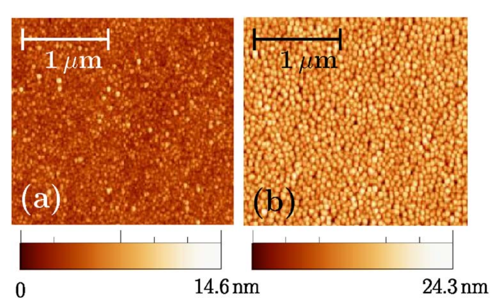

FIG. 4. (Color online) AFM topographies of C_{60} deposited on the BCB dielectric. (a) C_{60} grown on a nonpreheated BCB. (b) C_{60} grown on a *in situ* preheated BCB. Preheat time $t = 20$ min at 250 °C.

SCLC can increase the conductivity of wide-band-gap materials several orders in magnitude. A signature of SCLC for the OFET with C_{60} grown on the preheated dielectric, is the positive curvature in the output characteristics at high drain-source voltages (see Fig. 3). At low gate-source voltages $V_{GS} < 10$ V the space charge caused by the drain-source current (SCLC) dominates over the the gate induced space charge. In the range of $10 < V_{GS} < 40$ V the gate induced space charge dominates and a pinch off voltage followed by a saturated current is observed.[18] At higher gate-source voltages $V_{GS} > 40$ V the SCLC in the small depleted region increases the conductivity therein and the differential resistance in the saturation regime is reduced (e.g., at $V_{GS} = 50$ V no saturation of I_{DS} is observed at all). Further increase of V_{GS} caused a positive curvature of the output characteristics which develops into a sudden increase—*threshold*—of the drain-source current. We propose that the dramatic increase of I_{DS} for $V_{GS} \geqslant 70$ V is caused by trapped filled limited (TFL) current in the channel. Generally TFL currents are orders in magnitude higher than the corresponding trap-limited SCLC currents.[13] The transport under TFL conditions are no longer hindered by the presence of empty traps[19] and the effective mobility μ_{eff} is equal to the high microscopic value μ. In order to achieve TFL currents, high quality materials with a low total number of traps and a high electric fields is required. If the material has a mobility edge, the charge transport—under trap filled conditions—can be metallic.[20] Other effects during the preheating of the BCB dielectric, such as the removal of absorbed moisture or residual gases, are objects for further studies.

In summary, the growth parameters of the epitaxially grown C_{60} have a significant influence on the device performance of the OFET. OFET's with more amorphous C_{60} films feature a well defined pinch off voltage and a high differential resistance in the saturation regime. On the other hand, in OFET's containing a C_{60} film of enhanced crystallinity, a switching is observed with "on-currents" in the range of 100 mA. It is proposed that the switching is caused by space charge limited currents beside the gate induced space charge.

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[1] M. S. Dresselhaus, G. Dresselhaus, and P. C. Eklund, *Science of Fullerenes and Carbon Nanotubes* (Academic, New York, 1995).
[2] K. Tanigaki, I. Hirosawaand, T. Ebbesen, J. Mizuki, Y. Shimakawa, Y. Kubo, J. Tsai, and S. Kuroshima, Nature (London) **356**, 419 (1992).
[3] H. Sitter, D. Stifter, and T. N. Manh, J. Cryst. Growth **174**, 828 (1997).
[4] A. Andreev, G. Matt, C. J. Brabec, H. Sitter, D. Badt, H. Seyringer, and N. S. Sariciftci, Adv. Mater. (Weinheim, Ger.) **12**, 629 (2000).
[5] D. Stifter and H. Sitter, Appl. Phys. Lett. **66**, 679 (1995).
[6] H. Sitter, A. Andreev, G. Matt, and N. S. Sariciftci, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A **385**, 51 (2002).
[7] S. Sze, *Semiconductor Devices, Physics and Technology* (Wiley, New York, 2002).
[8] T. B. Singh, N. Marjanović, G. J. Matt, S. Günes, N. S. Sariciftci, A. M. Ramil, A. Andreev, H. Sitter, R. Schwödiauer, and S. Bauer, Org. Electron. **6**, 105 (2005).
[9] M. Koehler and I. Biaggio, Phys. Rev. B **70**, 045314 (2004).
[10] G. Horowitz and P. Delannoy, J. Appl. Phys. **70**, 469 (1991).
[11] C. Tanase, E. J. Meijer, P. W. M. Blom, and D. M. de Leeuw, Phys. Rev. Lett. **91**, 216601 (2003).
[12] A. Rose, Phys. Rev. **97**, 1538 (1955).
[13] M. A. Lampert and P. Mark, *Current Injection in Solids* (Academic, New York, 1970).
[14] F. Schauer, R. Novotny, and S. Nešpůrek, J. Appl. Phys. **81**, 1244 (1997).
[15] S. D. Baranovskii, H. Cordes, and F. Hensel, Phys. Rev. B **62**, 7934 (2000).
[16] G. J. Matt, T. Fromherz, and N. S. Sariciftci, Appl. Phys. Lett. **84**, 1570 (2004).
[17] R. W. de Boer, M. Jochemsen, T. M. Klapwijk, A. F. Morpurgo, J. Niemax, A. K. Tripathi, and J. Pflaum, Org. Electron. **6**, 105 (2005).
[18] B. H. Hamadani and D. Natelson, J. Appl. Phys. **97**, 064508 (2005).
[19] V. Podzorov, E. Menard, A. Borissov, V. Kiryukhin, J. A. Rogers, and M. E. Gershenson, Phys. Rev. Lett. **93**, 086602 (2004).
[20] N. Mott, Rev. Mod. Phys. **50**, 203 (1978).