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Strain induced anisotropic effect on electron mobility in C\textsubscript{60} based organic field effect transistors

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The electron mobility was found to increase (decrease) upon applied compressive (tensile) strain, respectively, when a high-performance flexible C\textsubscript{60}-based organic field-effect transistor (OFET) was subjected to different bending radii. The observed almost twofold relative change in the electron mobility is considerably larger than that reported before for pentacene-based OFETs. Moreover, the strain dependency of electron mobility in C\textsubscript{60} films is strongly anisotropic with respect to the strain direction measured relative to the current flow. Analysis within a hopping-transport model for OFET mobility suggests that the observed strain dependency on electron transport is dominated mostly by the change of inter-grain coupling in polycrystalline C\textsubscript{60} films. © 2012 American Institute of Physics.

Mechanical flexibility is one of the key advantages of organic materials in applications such as wearable-electronics or flexible displays.\textsuperscript{1-3} It is consequently important to understand the effect of strain on these materials due to rolling, twisting, and curling of large flexible substrates for a variety of applications. Curling of flexible substrate has been known to introduce strain in organic devices.\textsuperscript{2,4} In silicon-based integrated circuit applications, strain effects play a significant role in improving the device performance and considerable insight is also gained on the role of strain in charge carrier transport.\textsuperscript{5} In silicon, the effect of strain on p-type and n-type devices is complementary. It is reported that in silicon under compressive strain, the hole mobility in p-type devices increases whereas the electron mobility in n-type devices decreases. With tensile strain, the effect on carrier mobility for n- and p-type silicon devices is opposite in nature.\textsuperscript{5,6} Although silicon is crystalline and organic semiconductors (OSC) are polycrystalline and the impact of strain could be significantly different, it is interesting to compare the effect of strain in p-type and n-type OSC materials.

The effect of strain in p-type OSC like Pentacene was explored in detail by Sekitani et al.\textsuperscript{7} and Jedaa and Halik\textsuperscript{7} It is reported that on application of compressive strain mobility increases and under tensile strain it decreases. Furthermore, there are reports where strain is studied from point of reliability assessment and degradation where organic circuits are monitored with the application of strain to identify the bending limits of flexible devices.\textsuperscript{8} However, no systematic studies on effect of strain on n-type OSC have been reported in literature, which is the focus of this paper. We report the effect of strain on the n-type OSC C\textsubscript{60} that is well known for high performance organic field-effect transistors (OFETs).\textsuperscript{9,10} We have fabricated high mobility, flexible top gate C\textsubscript{60} transistors and systematically investigated the effect of bending on electronic charge-carrier transport.

For flexible substrates, we used 170\textmu m thick polyethylene terephthalate (PET) sheets. First, we spin coated a 1\textmu m thick divinyltetramethyl disiloxane-bis(benzocyclobutene) (BCB) as a smoothing layer to reduce the surface roughness of PET sheets. The substrate is then baked at 200°C to anneal the smoothing layer and to prevent shrinkage of sheets during further processing. Next, Al source-drain region of 70 nm thickness was deposited through evaporation. A 300 nm thick C\textsubscript{60} film was then deposited by hot wall epitaxy (HWE)\textsuperscript{9} technique. Subsequently, parylene-C was deposited as gate dielectric with a thickness of 1\textmu m, as explained in detail elsewhere.\textsuperscript{10} As a final step, the Al top gate was evaporated (70 nm). The top gate C\textsubscript{60} transistor on flexible PET substrate exhibited low leakage high-performance characteristics with a mobility of $\mu = 0.58\text{cm}^2/\text{Vs}$, no hysteresis, a threshold voltage $V_T = -0.1\text{V}$, and our on-off ratio of $10^5$. Figures 1(a) and 1(b) show the representative transfer and output characteristics of the device under consideration. All measurements were done in nitrogen atmosphere. Inset of Fig. 1(a) gives a typical AFM image of deposited C\textsubscript{60} film showing the polycrystalline morphology with estimated grain size of 80 nm. The polycrystalline nature of the films grown by HWE technique was also ensured by XRD measurements.\textsuperscript{11}

Strain was applied by curling the flexible substrate between screw clamps, as indicated in inset of Fig. 2, for different radius. The strain induced curvature of the flexible...
substrate was estimated by fitting the cross-sectional optical images. The strain $S$ applied to the OSC films was evaluated using an approximated formula, $S = D/2R$, where $D$ is the thickness of substrate and $R$ is the radius of curvature. Substrates were concavely and convexly curled in turn to apply the compressive strain and tensile strain, respectively. The measurements were done in two different configurations where applied strain is parallel or perpendicular to the direction of source-drain current in OFETs.

In the first set of experiments, we applied compressive and tensile strain and measured the current, parallel to the direction of applied strain. Fig. 2 shows the corresponding transfer characteristics. First, we measured the device at flat conditions and then reduced the bending radius $(15 \text{ mm} > R > 5.1 \text{ mm})$ in concave (compressive) direction. We then measured the device again in flat condition before bending it in convex (tensile) direction. The saturation current increases with increasing compressive strain and decreases with increasing tensile strain. For a bending radius of 5.1 mm, the saturation current increases by 80% and decreases by 70% for compressive and tensile strains, respectively, which is a major change in current levels. This change is found to be reversible after compressive strain but tensile strain caused a reduction in drain current by 65% after the first trial.

To further analyze the electron transport under strain condition, we have fabricated another set of devices with similar characteristics and measured the current with strain in perpendicular direction to source-drain current. For compressive strain, a small change in saturation current was observed but almost no change in case of tensile strain. The charge carrier mobility ($\mu$) is extracted from the slope of $\sqrt{I_D}$ versus $V_{GS}$ plot. In Figure 3, the change of electron mobility vs. strain for both set of experiments is depicted. Mobility monotonically increases (decreases) when the compressive (tensile) strain is applied parallel to direction of charge transport but the change is small and non monotonous for the perpendicular case. Mobility increased from 0.11 $\text{cm}^2/\text{Vs}$ to 0.22 $\text{cm}^2/\text{Vs}$, which is more than 100% of

![FIG. 1. (a) Transfer ($I_{DS}$-V$_{GS}$), (b) output ($I_{DS}$ vs. V$_{DS}$) characteristics, respectively, of high performance C$_{60}$ based flexible top gate OFET with W/L = 1800 $\mu$m/70 $\mu$m. $C_i$ = 2.6 nF/cm$^2$, $V_T$ = –0.1 V, $\mu$ = 0.58 cm$^2$/Vs, $I_{sat}/I_{off} = 10^3$. Inset of (a) gives 2 $\mu$m $\times$ 2 $\mu$m AFM image of the C$_{60}$ film along with the cross sectional device schematics.](image1)

![FIG. 2. Transfer characteristics $I_{DS}$-V$_{GS}$ when current is parallel to applied compressive and tensile strain. Inset gives the corresponding applied strain values and schematic of translation stage.](image2)

![FIG. 3. Mobility versus strain for a bending radius of R where 15 mm > R > 5.1 mm. Green and red curves correspond to parallel and perpendicular measurements, respectively. Inset shows four curling configurations pointing to mobility measurements in those states. Purple arrows depict the electron transport directions. Current parallel to (a) compressive and (b) tensile strain and current perpendicular to (c) compressive and (d) tensile strain. Mobility change with strain is more prominent for strain parallel transport to the transport than the perpendicular case.](image3)
increase, for 1.7% of compressive strain. This points towards the potential of C_{60} to be used for low cost stress sensor applications.\textsuperscript{13} The gate leakage of the devices was independent of strain which highlights the stability of parylene-C as a gate dielectric for bending applications.

The observed effect of compressive and tensile strain on electron transport in C_{60} films is qualitatively similar to that observed for p-type OSC like pentacene.\textsuperscript{4,7,13} However, as our result show, the change in mobility for C_{60} based devices is almost one order of magnitude stronger than that reported for pentacene.\textsuperscript{4,7} This may be due to the fact that our devices are top gate transistors where the dielectric layer acts as an encapsulation layer. The top encapsulation layer supports the device performance. It reduces the probability of grains being dislocated and ensures a homogenous distribution of force in the OSC layer.

A remarkable result of this study is that the strain effect on mobility in C_{60}-based FETs is found to be strongly anisotropic with respect to the direction of applied strain. This observation is in variance to earlier observations by other research groups in pentacene OFETs where the change in source-drain current did not depend on direction of strain relative to the direction of current flow.\textsuperscript{4} The change of $\mu$ upon compressive or tensile strain can, in principle, be either due to the change of the intermolecular distances affecting the overlap between neighboring sites or/and due to the change of the energy distribution of localized sites (energetic disorder) in the film. It is worth noting that an increase in conductivity under the applied pressure has been demonstrated in OSC films long ago\textsuperscript{14} and was attributed to the enhanced intermolecular coupling in films. Applied strain could also change the energy of localized states (traps) controlling the charge carrier mobility because in non-polar material like C_{60}, the electronic polarization energy is mainly determined by intermolecular distances.\textsuperscript{14} However, the creation of deeper localized states (or traps) will only be determined by the stress magnitude and should not depend on the applied stress direction. Thus, the observed strong anisotropy of strain effect implies that the change in the charge mobility in C_{60} films is mostly dominated by the intermolecular coupling factor rather than the energy distribution of traps, and thus it can discriminate between these factors.

To get a deeper insight into the origin of the observed strain effect on the charge mobility, we fitted our experimental data by the Fischchuk analytic extended gaussian disorder (EGD) model for OFET mobility,\textsuperscript{15,16} which has been recently used to describe temperature activated- and electric field dependent OFET mobility in C_{60} films. All material parameters used were the same as for bottom-gate C_{60} OFET studied before\textsuperscript{15} and only the hopping distance between neighboring sites, $a$, was varied to get the best fit with the experimental data. For films in flat condition, we used $a = 1.4 \text{ nm}$ (intermolecular distance in C_{60} crystals)\textsuperscript{15} as a representative value for such films. The parameter $a$ affects mostly the mobility prefactor, the position of the effective transport energy level and the electric-field dependence term (see Ref. \textsuperscript{16} for details). Mobilities calculated according to Ref. \textsuperscript{16} as function of hopping distance are presented in Fig. 4 (red solid curve) and demonstrate a good agreement with the experimental data (symbols) on the strain dependence of $\mu$, especially for strain values less than 1%. Some deviation of the mobility experimental data from the calculated dependence might be due to a film adhesion problem at large enough strains. Similar fitting results were also obtained by the Pasveer/Coehoorn EGD model\textsuperscript{18} using the same set of material parameters (blue solid curve in Fig. 4).

An essential conclusion that can be drawn from the comparison of experimental results and modeling is that in order to explain the observed twofold increase (decrease) of $\mu$ upon applied 1.7% compressive (tensile) strain in C_{60} films, one has to assume a relative decrease (increase) in intermolecular distance as large as $\sim 10\%$. The latter is clearly impossible for the 1.7% strain because even for ideal adhesion between substrate and C_{60} films, the change of the intermolecular distance under such a condition is expected be of the order of 1%-2% only.

To solve the puzzle, one has to take into account the fact that thin films of C_{60} have polycrystalline\textsuperscript{19} morphology with a number of grains and grain boundaries. Grain boundaries form the weakest link in organic thin films in terms of mechanical deformation; it is, therefore, logical to assume that the majority of applied strain will be absorbed at the grain boundaries. When strain is applied, these grain boundaries tend to get closer or move apart leading to a change in grain boundary resistance.\textsuperscript{20,21} This change in resistance is captured as a change in mobility, as observed in our results. It also explains the anisotropic effect of strain on mobility, as observed in our results. In the case of perpendicular strain, grain boundaries perpendicular to conduction get closer but they will have lesser impact on the transport.

Thus, we suggest that relative change of inter-grain distances (the effective hopping distance) can be quite considerable and is responsible for the observed strain effect on the OFET charge mobility. First, grains are weaker coupled between each other compared to intermolecular coupling within crystallites. Second, it is well-known that charge transport in polycrystalline films is controlled by grain
boundaries which create major potential barriers\textsuperscript{21–23} (both traps and scattering centers) between the more ordered domains. Indeed, the mobility activation energy in C\textsubscript{60} films was recently shown to decrease with increasing average grain size\textsuperscript{24} accompanying by drastic enhancement of the electron mobility and the grain boundaries were recently found to determine the electric field dependence of $\mu$ in polycrystalline OSC films.\textsuperscript{22} As we demonstrated recently,\textsuperscript{22} the rate limiting charge transfer events are likely to be the inter-grain jumps; therefore, the complex charge hopping transport problem in such films could be reduced to considering just the most difficult jumps over the grain boundaries. This is reminiscent of charge transport in a conjugated polymer where the limiting step is the jump between conjugated (more ordered) segments of neighboring chains. The barrier heights due to grain boundaries are subject to distribution over the film, therefore taking into account a huge variety of percolative passes present between the wide (1.8 mm) source and drain electrodes of an OFET device, the charge transport in average could be considered as that occurring in an effectively random disordered system even though charge carriers may experience just a few crossings over grain boundaries in a particular percolative pass. Thus, it is plausible that the energy distribution of the charge donating and charge accepting states at the grain boundaries follows a Gaussian distribution. This justifies the application of the Gaussian disorder model to consider the average charge mobility in such OFET channels. This idea has been recently verified experimentally for polycrystalline silylethynyl-substituted pentacene films\textsuperscript{22} and thereby the EGD model is able to accommodate grain boundaries to describe charge transport in polycrystalline OSC films.

In conclusion, strain effect on electron transport was studied in top-gate flexible C\textsubscript{60}-based OFET devices fabricated on PET substrate and using a parylene-C as gate dielectric. Change in the electron mobility upon applied 1.7% of compressive/tensile strain is found to be relatively large considerably exceeding that reported before for other OFETs. In contrast to the latter, the strain influence of electron mobility in C\textsubscript{60} films is strongly anisotropic with respect to the direction of applied strain. The observed anisotropy suggests that the effect is dominated by the change of the effective hopping distance—most probably of jump distances over the grain boundaries which dominate the charge transport in polycrystalline C\textsubscript{60} films. Furthermore, in roll to roll processing, for better strain reliability and for sensor applications, the direction of current should be made parallel and perpendicular to major rolling axis, respectively.

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