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Description	

Fabrication and characterization of C₆₀ thin-film transistors with high field-effect mobility

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We report an improvement in performance of C₆₀ thin-film field-effect transistors (TFTs) fabricated by molecular-beam deposition. Devices, fabricated and characterized under a high vacuum without exposure to air, routinely showed current on/off ratios $> 10^8$ and field-effect mobilities in the range of 0.5–0.3 cm²/V s. The mobility obtained is close to that derived from the photocurrent measurements on C₆₀ thin films and comparable to a very high value among *n*-type organic TFTs.

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Performances of organic semiconductors are often measured by the field-effect mobility. 1 cm²/V s and on/off ratios in the range of 10⁶ are conventionally used as criteria because amorphous silicon, used as drivers of liquid-crystal displays, typically shows these features.¹ Among a variety of organic semiconductors, pentacene has been known as the best material because of its high mobility of 1.5 cm²/V s or more, which was achieved using a SiO₂ surface treatment technique with organic monolayers.² Since pentacene thin-film field-effect transistors (TFTs) usually show *p*-type operation, researchers have searched for *n*-type materials with high mobility for making complementary circuits. Though high performance TFTs with *n*-type materials are highly demanded, the mobilities of *n*-type TFTs remained relatively low.¹ Fullerene C₆₀ has been known as an *n*-type semiconductor with the highest mobility of 0.08 cm²/V s and 0.3 cm²/V s for untreated and treated with tetradiaminoethylene molecules, respectively.³ Recently, mobilities of fluorinated phthalocyanine,⁴ naphthalene, and perylene derivatives, such as N-substituted naphthalene 1,4,5,8-tetracarboxylic diimide⁵ or N,N'-dialkyl-3,4,9,10-perylene tetracarboxylic diimide (PTCDI) has been found to be a promising group of materials. In 2002, mobilities up to 0.6 cm²/V s and current on/off ratio $> 10^5$ were reported in PTCDI-C8H.⁶

Among the *n*-type molecular materials, the room-temperature mobility of a pure C₆₀ TFT was extremely high among organic semiconductors in 1995. However, to date, there is no improvement of mobility in C₆₀ TFTs.^{3,7} This is predominantly because of the extreme sensitivity to air exposure of C₆₀ TFTs, which is more or less a generic feature of *n*-type TFTs. Once C₆₀ TFTs are exposed to air, the performance is completely destroyed. Though evacuation recov-

ers the performance, the mobility of 0.1 cm²/V s, derived by the *in situ* measurement, has not been overcome by *ex situ* measurements. However, recent developments in the passivation technique showed that a C₆₀ TFT operated even in air.⁷ Hence, the intrinsic device performance of C₆₀ should be worth revisiting. In this letter, we report the performance of C₆₀ TFTs which were grown by molecular-beam deposition. The mobility up to 0.5 cm²/V s and on/off ratios $> 10^8$ were obtained by an *in situ* measurement in a high vacuum chamber. The high value of mobility, 0.56 cm²/V s, is comparable to that of single crystal C₆₀, determined by the time-of-flight experiment.⁸ Atomic force microscope (AFM) images of the C₆₀ film revealed that the grain size increases with increasing substrate temperature, while the mobility did not exhibit a clear relation with the substrate temperature. The present result indicates that grain size is not very crucial for high mobility in C₆₀ TFTs, in contrast to organic TFTs with planar molecules.

We have fabricated C₆₀ TFTs with a bottom contact configuration. A heavily doped *p*-type silicon wafer was used for a back gate electrode with a 400 nm insulating layer of thermally grown silicon dioxide on top of it. 10/100 nm Ti/Au or Cr/Au drain and source electrodes were fabricated on the surface of SiO₂ layer, using photolithography and lift off. The devices had a channel length (*L*) of 50 μm and a width (*W*) of 2 mm. The substrate was annealed at 150 °C under 1×10^{-9} Torr for 10 h before the deposition of C₆₀. Except for this annealing, no other surface treatment was made on the silicon dioxide insulator. C₆₀ films of 150 nm in thickness were grown by molecular-beam deposition at the base pressure below 10^{-9} Torr during deposition with the rate of 0.15 Å/s. The substrate temperature (*T_S*) was kept constant during deposition of C₆₀ from a Knudsen cell. *T_S* was changed from room temperature to 130 °C. After the deposition of the C₆₀ film, the substrate was cooled down to room temperature over a few hours and the device was transferred to a charac-

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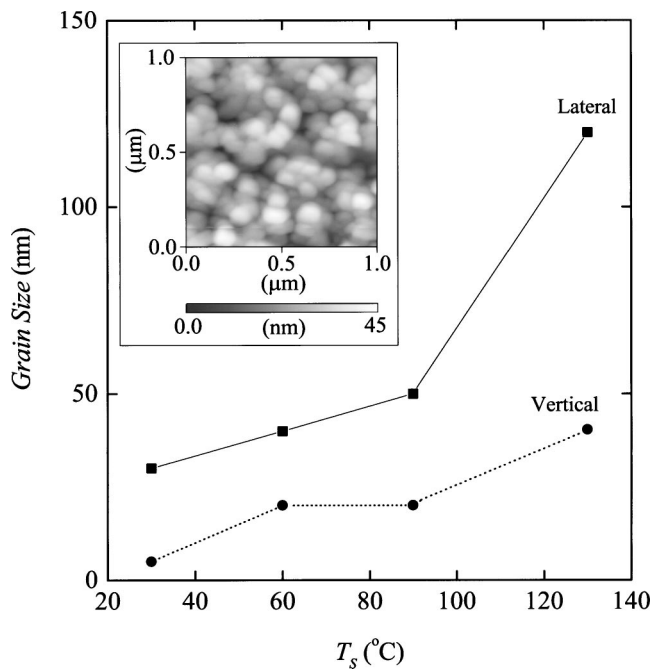


FIG. 1. Correlation between grain sizes and substrate temperature T_S . The grain size for lateral and vertical directions were estimated from the AFM images. The inset shows the AFM image of the C_{60} TFT device fabricated at $T_S = 130$ °C.

terization chamber kept at 5×10^{-10} Torr without exposing to air. This additional characterization chamber enables us to have an effective and quick characterization of devices. The device properties were measured by three current/voltage probes equipped with the high vacuum chamber.

We have tested the effect of covering SiO_2 surface with monolayers of organic molecules such as hexamethyldisilazane (HMDS), which is well known to minimize the interaction between organic molecules and SiO_2 substrates. HMDS was not very effective for improving the field-effect mobility of C_{60} .

The crystallinity and morphology of the C_{60} thin films were characterized by means of electron diffraction, x-ray diffraction, and AFM. Figure 1 shows the variation of average grain size determined from AFM images (shown in the inset of Fig. 1) against substrate temperature T_S . The grain size increases with T_S both for the lateral and vertical directions. Both x-ray and electron diffraction experiments showed three intense Debye-Scherrer rings indexed as (111), (220), and (311) reflections of face-centered-cubic lattice with the cell parameter of 14.1 Å, indicating that films are polycrystalline without notable orientations. The size of crystallites calculated from the full width of half maximum of the x-ray diffraction peaks was about 10 nm. This value of crystallite size is smaller than the grain size estimated from the AFM images, indicating that the grains shown in Fig. 1 are composed of several crystallites.

Figure 2 displays a three-dimensional plot of drain current I_D as a function of drain voltage (V_D) and gate voltage (V_G). I_D increases almost linearly with V_D followed by a saturation due to the pinch off of the accumulation layer. Figure 3 shows a relation between I_D and V_G at $V_D = 100$ V for the same device. The left- and right-hand side vertical axes show the logarithm and root of I_D , respec-

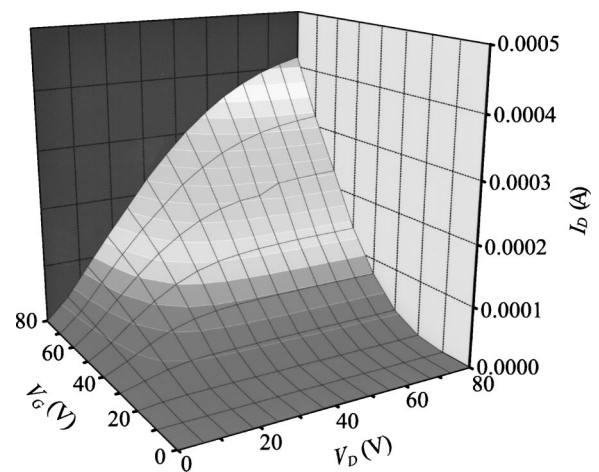


FIG. 2. Three-dimensional plot of I_D as a function of V_D and V_G for C_{60} TFT with a bottom gate configuration measured at room temperature at 5×10^{-10} Torr. $L = 50$ μm and $W = 2$ mm.

tively. The left-hand side scale indicates that the on-off ratio larger than 10^8 is achieved in this device. From the linear fit of a $I_{SD}^{1/2}$ versus V_G plot in the saturation region, the field-effect mobility $\mu = 0.56$ cm²/V s and the threshold voltage $V_{th} = 17$ V were derived assuming the capacitance of 1.0×10^{-8} F/cm². The mobility between 0.5 and 0.3 was routinely obtained in the present method. These values are comparable to the electron mobilities determined by the time-of-flight experiment carriers on C_{60} single crystals⁸ or by the photocurrent measurements on C_{60} thin films.⁹ Furthermore, this value is close to the maximum mobility 0.6 cm²/V s among the n -type organic TFTs, which is reported for PTCDI-C8H.

Interestingly, the mobility did not display a strong correlation with the substrate temperature, implying that the mobility does not strongly depend on the grain size determined from AFM. This result is in sharp contrast with other planar molecules. For instance, it is well known that the mobility of pentacene TFT is strongly dependent on the orientation and domain size of crystallites in the film.¹ The present results, on the other hand, indicate that grain size is not crucial for high mobility in C_{60} , at least at room temperature.

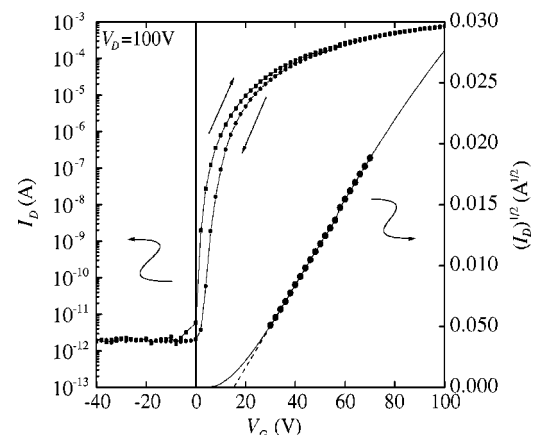


FIG. 3. Semilogarithmic plot of I_D vs V_G (left-hand side scale) and plot of $I_D^{1/2}$ vs V_G (right-hand side scale) for the same C_{60} TFT device as in Fig. 2. The dashed line shows a fit to the equation in the saturation region, $I_D = WC\mu/2L(V_G - V_{th})^2$, from which field-effect mobility μ was obtained as 0.56 cm²/V s. Here, $C = 1.0 \times 10^{-8}$ F/cm² is a capacitance of the device.

The mobility of a C₆₀ TFT seems much more sensitive to the atmosphere. In fact, a high vacuum, as well as long time evacuation of chamber, was important for the present improvement of mobility of C₆₀ in the present experiment. Also, we frequently encountered the increase in mobility by a factor of 2 by overnight pumping after deposition. These results indicate that a high vacuum or inert atmosphere, rather than the grain size, is important for the high mobility of C₆₀ TFTs. Furthermore, the recent finding of passivation by covering a C₆₀ TFT with Al₂O₃ is useful to keep the high performance of the device achieved in a high vacuum.⁷ These results indicate that C₆₀ is promising as an *n*-type high mobility device.

In summary, we have shown that the *n*-type high mobility of 0.5 cm²/V s is routinely achieved in C₆₀ TFTs fabricated and characterized in a high vacuum. The obtained mobility is comparable to that of single crystals determined by time-of-flight experiment or to that of *n*-type PTCDI-C8H. The high vacuum seems to be crucial for the high mobility, rather than the grain size.

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