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Description	

Fabrication and characteristics of C_{84} fullerene field-effect transistors

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Fullerene field-effect transistors (FETs) were fabricated with thin films of C_{84}, which showed n-channel normally-on depletion-type FET characteristics. The C_{84} FET device exhibited the highest mobility, μ, of 2.1×10^{-3} $cm^2\,V^{-1}\,s^{-1}$ among normally-on fullerene FETs. The carrier transport of this FET device can be interpreted as thermally activated hopping transport. Carrier type (n-channel) and transport mechanism (hopping) reflect the electronic properties of the C_{84} molecule. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1695193]

A large number of field-effect transistors (FETs) with thin films of organic molecules have been fabricated and their characteristics have been studied for next-generation electronics during the past decade.[1,2] The field-effect mobilities μ of organic FETs are lower by four orders of magnitude than those of conventional FETs with inorganic materials. Nevertheless, organic FETs are known to have many advantages such as large-area coverage, structural flexibility, and low-temperature and low-cost processing in comparison with inorganic FETs. In 1997 the mobility of a pentacene p-channel FET reached $1.5\,cm^2\,V^{-1}\,s^{-1}$.[3] Subsequently, a p-channel FET on single crystals of rubrene showed a mobility of $1.0\,cm^2\,V^{-1}\,s^{-1}$.[4]

The first fullerene FET was fabricated with thin films of C_{60} by Haddon *et al.*[5] This FET device showed a high mobility of $0.08-0.3\,cm^2\,V^{-1}\,s^{-1}$. The high μ value of this device implies that C_{60} FETs can play an important role in future applications such as identification tags, smart cards, and drivers for active-matrix displays based on the integration with organic light-emitting diodes. The mobility values of n-channel organic FETs except for the C_{60} FET are much lower than those of p-channel organic FETs.[1,2] Furthermore, such high μ value attracts interest in the physics and chemistry of C_{60} FETs. The improvement of properties of the C_{60} FET device has long been examined, and very recently the mobility reached $0.56\,cm^2\,V^{-1}\,s^{-1}$, which is one of the highest mobilities among n-channel organic FETs.[6] The highest μ of $0.6\,cm^2\,V^{-1}\,s^{-1}$ was reported for N,N'-dialkyl-3,4,9,10-perylene tetracalboxylic diimide derivative (PTCDI-C8H) FET device.[7]

Recently we reported the transport properties of C_{60} FET.[8] Temperature dependence of the mobility suggested a hopping transport as a conduction mechanism for these FETs. Furthermore, a complementary metal–oxide–semiconductor logic gate circuit was fabricated with C_{60} and pentacene FETs,[8] which leads us to realize various types of logic circuits such as NOR and NAND for computing and memory. The first FETs with metallofullerenes have been realized with $Dy@C_{82}$ and $La_2@C_{80}$.[8,9] These FETs operated as n-channel normally-on device, which is substantially different from enhancement-type FETs with C_{60} and C_{70}.[5,6,8,10] The output characteristics of fullerene FETs, therefore, reflect intrinsic electronic structures of individual fullerene molecules. This letter reports on the fabrication of higher-fullerene FET devices with thin films of C_{84}, and the FET characteristics and their temperature dependence.

Schematic representation of C_{84} (D_{2d} isomer) and a cross-sectional view of the C_{84} FET device are shown in Fig. 1(a). Commercially available C_{84} (99%) was used for the fabrication of the thin film. Commercially available $SiO_2/Si(100)$ wafers were used as substrates after cleaning with acetone, methanol, and H_2SO_4/H_2O_2. The thin film of C_{84} was formed by a thermal deposition under a vacuum of 10^{-8} Torr. The channel length L and the channel width W of this device were 75 and 4000 μm, respectively. The characteristics of the C_{84} FET device were measured under 10^{-6} Torr after annealing for 24 h at 120 °C under 10^{-6} Torr.

The drain current I_D versus drain–source voltage V_{DS} plots for the C_{84} FET at 290 K are shown in Fig. 1(b). The

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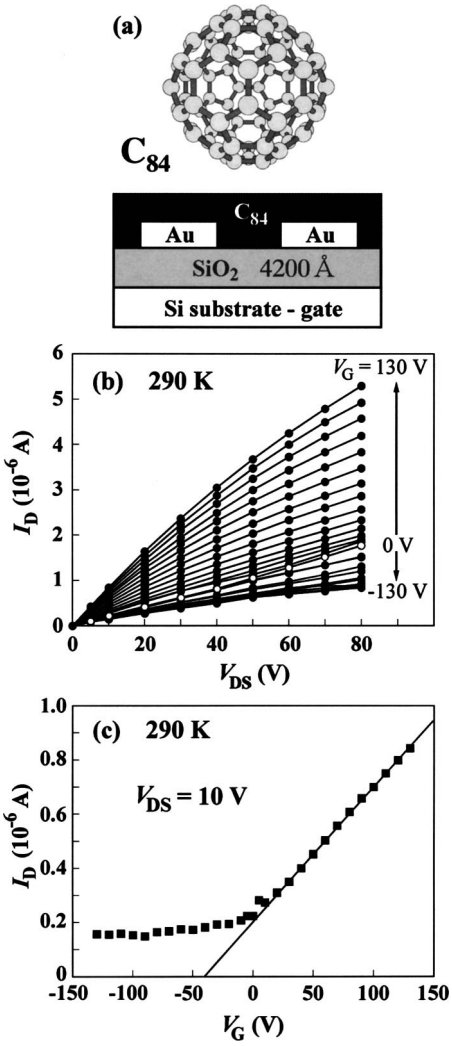

FIG. 1. (a) Schematic picture of $C_{84}(D_{2d})$ and cross-sectional view of C_{84} FET. (b) I_D-V_{DS} plots of C_{84} FET at 290 K. Closed and open circles refer to the points measured at $V_G \neq 0$ V and $V_G = 0$ V, respectively. (c) I_D-V_G plot at $V_{DS} = 10$ V. Solid line is the line fitted with equation $I_D = (\mu W C_0/L)(V_G - V_T)V_{DS}$, where C_0 refers to capacitance of SiO_2; $C_0 = 8.22\times10^{-9}$ F cm^{-2}.

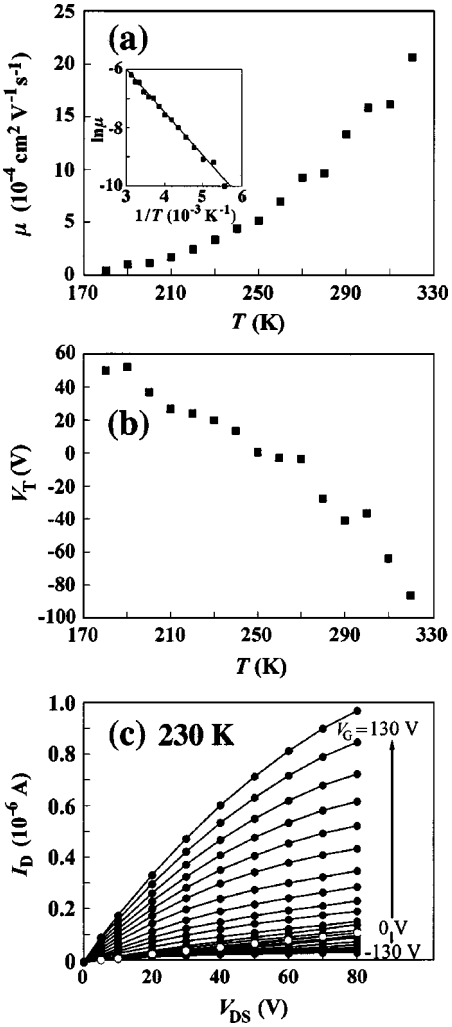

FIG. 2. (a) $\mu-T$ plot of C_{84} FET and ln μ-(1/T) plot in the inset. Solid line is the line fitted with the equation ln $\mu \sim E_a/k_BT$, where k_B is Boltzmann constant. (b) V_T-T plot of C_{84} FET. (c) I_D-V_{DS} plots of C_{84} FET at 230 K. Closed and open circles refer to the points measured at $V_G \neq 0$ V and $V_G = 0$ V, respectively.

plots show output characteristics of n-channel normally-on depletion-type FET. Relatively large I_D is observed even at zero gate voltage V_G. By varying V_G from -130 to 130 V, I_D increases as V_G increases as shown in Fig. 1(b). It can be seen that the channel conductance decreases by applying a negative V_G, presumably due to the depletion of carriers in the channel. I_D remained constant below $V_G = -70$ V. This can be explained by assuming the existence of bulk current which cannot be reduced by applying the negative V_G. The threshold voltage V_T was estimated to be -42 V from the I_D-V_G plot at $V_{DS} = 10$ V [Fig. 1(c)]. The negative V_T supports that the C_{84} FET is normally-on type. Very recently such a normally-on depletion-type property was observed in $La_2@C_{80}$ FET.[9]

The field-effect mobility for the C_{84} FET was estimated to be 1.1×10^{-3} cm^2 V^{-1} s^{-1} from the I_D-V_G plot [Fig. 1(c)]. This value is higher by one order of magnitude than those reported for $Dy@C_{82}$ and $La_2@C_{80}$ FETs.[8,9] This implies that C_{84} is superior to metallofullerenes such as $Dy@C_{82}$ and $La_2@C_{80}$ as materials for an active layer in normally-on depletion-type FET. The μ increases monotonically as temperature increases up to 320 K [Fig. 2(a)]; the mobility reached 2.1×10^{-3} cm^2 V^{-1} s^{-1} at 320 K. FET behavior was not clearly observed below 180 K because of very small I_D. The activation energy E_a was estimated to be 0.13 eV from the ln μ versus T^{-1} plot shown in the inset of Fig. 2(a). These results show that the channel conduction of the C_{84} FET device follows a thermally activated hopping-transport model ($\mu \sim \exp - E_a/k_BT$). One of the origins of high μ in the C_{84} FET may be attributed to the fact that the channel conduction occurs through a hopping between the delocalized lowest unoccupied molecular orbitals (LUMOs) in C_{84}, which is different from that between the localized LUMOs dominated by encapsulated metal ions in metallofullerenes.[9]

The threshold voltage V_T decreases monotonically with increasing temperature up to 320 K [Fig. 2(b)]. Such temperature dependence might be caused by an increase in bulk current. Actually the bulk current increased with increasing temperature,[11] which results in an apparent variation in V_T. The V_T values are positive at temperatures below 250 K, and the FET property apparently changes from the normally-on to a normally-off with decreasing temperature. As an ex-

ample of the normally-off like FET, the $I_D - V_{DS}$ plots at 230 K are shown in Fig. 2(c).

The high I_D at $V_G = 0$ V in the high-temperature region can be explained by a small gap energy, E_g, of ~ 0.55 eV which was estimated from the temperature dependence of resistivity ρ for the C_{84} thin-film ($V_G = 0$ V).[11] The E_g value of the C_{84} thin film is much smaller than those determined for C_{60} (1.8 or 2.1 eV) and C_{70} (2.2 eV).[12–14] The C_{60} and C_{70} FETs showed enhancement-type normally-off properties.[5,6,8,10] Consequently, the normally-on type FET property in the C_{84} FET device can be interpreted within the framework of high bulk current in the C_{84} thin film. However, the origin of carriers of the high bulk current in the C_{84} thin film remains to be clarified. The on–off ratio, I_D ($V_G = 130$ V)/I_D ($V_G = -130$ V), at $V_{DS} = 10$ V of the C_{84} FET was ~ 6 at 290 K, as shown in Fig. 1(b). This low on–off ratio is owing to high bulk current of the C_{84} thin film. On the other hand, the on–off ratio increases at low temperatures, and the value reached ~ 37 at 230 K [Fig. 2(c)]. This increment is caused by the reduction of bulk current at low temperature. This will open a way to fabricate practical devices with higher fullerene FETs for computing and memory.

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