Carrier Injection Property of C\textsubscript{60} Thin Film Transistor via Displacement Current Measurement

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Abstract. Carrier injection into a C\textsubscript{60} thin film has been studied from characteristics of a field effect transistor and a displacement current measurement (DCM) using three kinds of metals, Mg, Ag, and Au. The Mg sample shows a highest field effect mobility of 0.62 cm\textsuperscript{2}/V s and a lowest threshold voltage of 6 V among the samples. A carrier injection and the accumulation are confirmed in the DCM curves by sweeping voltage applied on the back gate. The electron number and the density injected from the electrodes are estimated from the integration of the DCM curves. However, only the curve of the Mg sample shows a two-stage transition in the DCM curves. It could be due to a diffusion of Mg atoms into the C\textsubscript{60} layer and a transition of releasing electrons from the donor level could be observed at lower voltage region.

1. Introduction
Recently, organic semiconductors are getting applied for light emitting devices, and they are also expected to be applied for their drive transistors. However the operation mechanism, especially the origin of the carrier accumulation or injection, is not yet clear [1]. In n-type organic materials, C\textsubscript{60} is known to show relatively high mobility [2, 3] therefore an appropriate metal electrode is desired to form a contact with a low injection barrier for further improvement of mobility for its application. Because of n-type FET, low-work-function metals are expected to decrease the injection barrier for C\textsubscript{60} and to improve the mobility. For example, Eu, whose work function is smaller than the LUMO level of C\textsubscript{60}, has been tried to form a FET [4]. We have, therefore, studied a carrier injection property of C\textsubscript{60} thin film transistor using displacement current measurement (DCM), which has been used to characterize a carrier accumulation into an organic layer [5]. In this paper, we have confirmed carrier injection characteristics of three kinds of metal, Mg, Ag and Au, by the FET operation and the DCM with changing the measuring temperatures.

2. Sample and experimental setup
C\textsubscript{60} thin films were deposited 100 nm by thermal evaporation on a thermally grown SiO\textsubscript{2} layer on a p\textsuperscript{++}-Si substrate which was used as a back gate. Three kinds of metals, Mg, Ag and Au, were used for the electrical contact and were deposited on the C\textsubscript{60} thin films forming a bottom contact structure. In cases of the Mg and the Ag contact, Au thin film was deposited on the electrodes to avoid the oxidation. The samples were measured in vacuum of \textasciitilde10\textsuperscript{-6} torr after annealing at 430 K for 24 hours. For a determination of the FET characteristic, current across the source and drain (I\textsubscript{SD}) was measured with changing the back gate voltage (V\textsubscript{bg}). For the DCM, displacement current (I\textsubscript{d}) was measured
between the electrodes on the top of the C\textsubscript{60} layer (the source and the drain electrodes) and the back gate by sweeping the $V_{bg}$ with a rate of 1 mV/s.

### 3. Results and discussion

$V_{bg}$ dependences of the square root of $I_{SD}$ are shown in Fig. 1 with difference contact metals. All of the FETs show $n$- and enhancement type behaviour. The field effect mobility ($\mu$), the threshold voltage and ($V_{th}$) of the DCM curve are shown in Table 1. The $V_{th}$ in the Mg sample shows the smallest value of 6 V among these samples. It would be due to the smallest difference of energy between the LUMO level of C\textsubscript{60} (3.6 eV) and the work function of the metal (3.6 eV for Mg, 4.3 eV for Ag, and 4.9 eV for Au). Consequently, the $\mu$ in Mg sample shows the highest value of 0.62 cm$^2$/Vs. It is confirmed that lowering of the barrier height at the interface of C\textsubscript{60} and the metal is advantageous for increase of the mobility in a C\textsubscript{60} based FET.

![Figure 1. $I_{SD}$-$V_{bg}$ characteristics of the FET structures with Mg (circle), Ag (square) and Au (triangle) as the contact metals. Square root of $I_{SD}$ is used for the vertical axis to determine the $V_{th}$. The $V_{th}$ in Mg, Ag and Au samples are 6, 10 and 11 V, respectively.](image)

DCM curves in Mg, Ag and Au samples are shown in Figs. 2(a)-2(c), respectively. In the negative $V_{bg}$ region, no carrier is injected into the C\textsubscript{60} channel. Therefore, the C\textsubscript{60} layer acts as an insulating layer and forms a series connection of the capacitances. The $I_d$ can be considered to flow across the series capacitances of insulator layers of the C\textsubscript{60} and the SiO\textsubscript{2} and would be expressed by the following equation,

$$I_d = (S_s + S_d) \frac{C_c C_s}{C_c + C_s} \frac{dV_{bg}}{dt},$$  \hspace{1cm} (1)

where $S_s$ and $S_d$ are the areas of the source and drain electrodes, respectively, and $C_c$ and $C_s$ are the capacitance of C\textsubscript{60} and SiO\textsubscript{2} layers, respectively. The $I_d$ can be calculated as 0.021 nA, which is indicated by broken line (i) in Fig. 2(a), from the practical values and it is very consistent with the $I_d$ value at the left side of the DCM curve as shown in Fig. 2(a). On the other hand, when $V_{bg}$ exceeds the $V_{th}$, the electrons begin to be injected from the metal electrodes and to be accumulated at the interface between the SiO\textsubscript{2} and C\textsubscript{60} thin film. At enough positive $V_{bg}$, the $I_d$ would be consequently expressed by the following equation,

$$I_d = S_c C_s \frac{dV_{bg}}{dt},$$  \hspace{1cm} (2)

where $S_c$ is the areas of the C\textsubscript{60} thin film. These scenarios are well applicable at the right hand of the DCM curve at enough positive $V_{bg}$. One of the features of these DCM curves of C\textsubscript{60} thin films is a peak of the $I_d$ just before the electron widely distributing over the C\textsubscript{60} layer. The voltage which is
peak does not appear when the carriers are going to be depleted by sweeping the $V_{bg}$ to the negative direction. Considering the sweep direction, it would be due to a difference of efficiency of the barrier shape for carrier injection at the interface. Because the carriers are injected into the C$_{60}$ layer by tunnelling the barrier in a regime of Fowler-Nordheim-type field emission in the positive sweep [6]. A sudden increase of carrier injection could cause an excess current and show a peak in the DCM curve. Such a peak was observed also in a DCM curve of a pentacene FET [5]. On the other hand, thermionic emission would be dominant when the carrier is going back to the metal region. The carriers could be continuously returned to the metal region hence no peak would appear in this regime. The peak voltage ($V_p$) of the DCM curve are different in each sample as shown in Table 1. Nevertheless, it can be confirmed that a trend of the voltage shift of the $V_p$ is similar to that of the $V_{th}$.

The injected carrier (electron) number and the density can be estimated by integrating the DCM curve,

$$ Q = \int_{t_0}^{t_1} [I_d(t) - I_{d0}] dt, $$

where $I_{d0}$ is displacement current before starting the injection of carriers into the C$_{60}$ layer, and $t_0$ and $t_1$ are the times when the slope of triangle wave of $V_{bg}$ is inverted. For the Mg sample, the amount of the injected carrier and the concentration were estimated $3.5 \times 10^{-8} \text{ C}$ and $1.6 \times 10^{12} /\text{cm}^2$, respectively, by integrating the area colored by gray in Fig. 2(a). In the same way, the values in Ag and Au samples are also summarised in Table 1.

Figure 2. DCM curves of the C$_{60}$ thin films with contacts of Mg (a), Ag (b) and Au (c) electrodes observed at room temperature. (i) and (ii) in Fig.2(a) show 0.021 nA and 1.8 nA, respectively. The area painted by gray is used for the estimation of the amount of injected carriers.
Table 1. FET and DCM characteristics of each sample with different contact metals.

| Contact | $\mu$ (cm²/Vs) | $V_{th}$ (V) | $V_p$ (V) | Injected electron a | Electron concentration a |
|---------|----------------|--------------|-----------|---------------------|-------------------------|
| Mg      | 0.62           | 6            | 9.5       | $3.5 \times 10^{-8}$ | $1.6 \times 10^{12}$/cm² |
| Ag      | 0.34           | 10           | 21.5      | $2.6 \times 10^{-8}$ | $8.8 \times 10^{11}$/cm² |
| Au      | 0.076          | 11           | 22        | $2.0 \times 10^{-8}$ | $6.9 \times 10^{11}$/cm² |

aThese are estimated from integral of the displacement current curve and the area of the C₆₀ thin film.

Another feature can be seen in the Mg sample that there is a shoulder before the peak in the DCM curve as shown in Fig. 2(a). A similar shoulder is observed also in the back sweep of $V_{bg}$. Such a clear two-stage transition cannot be observed in the curves of the Ag and the Au sample at room temperature as shown in Figs. 2(b) and 2(c). In order to clarify the mechanism, the DCM curves of the Mg sample are measured at different temperatures as shown in Fig. 3. By decreasing temperature, the curves shift to the right (high $V_{bg}$) direction, consequently the shoulders of the lower transition also shift to the right. On the other hand, the $I_d$ value at the shoulder decays with lowering temperature and the structure almost disappears at temperatures lower than 220 K. Comparing the metals, it is well known that alkali- and alkali-earth metals are easy to diffuse into the C₆₀ crystal and act as donor [7, 8]. Therefore, if we assume that Mg atoms have diffused into the C₆₀ layer and a doped layer would extend underneath the Mg electrode. In this scenario, Mg atoms form a certain impurity level under the LUMO level and then the electron can be donated from the Mg atoms with much smaller electric field at higher temperatures. However, at lower temperatures below 220 K, the thermal energy would not be sufficient to release electrons and only the injected electron from the electrode would be dominant to accumulate in the C₆₀ layer. Therefore, such a two-stage transition could not be observed at lower temperatures. However, further study is necessary to clarify the difference.

Figure 3. DCM curves of the Mg sample measured at different temperatures. The curves are magnified around the first transition. The broken lines are guide for eyes to understand the shift of the shoulders by decreasing of temperature.
4. Summary
Carrier injection into a C_{60} thin film has been studied from the FET and the DCM characteristics in samples with contacts of Mg, Ag and Au. The Mg sample shows the highest mobility and the lowest V_{th} among the samples. A peak of displacement current is observed in the DCM curves of all the samples. The V_{p} shifts with the same trend of the V_{in} depending on the contact metals. Two-stage transition is observed only in the DCM curve of the Mg sample. It is confirmed that the DCM curve shifts to higher V_{bg} direction and the lower transition is smeared by decreasing the temperature. The transition would be caused by a diffusion of Mg atoms into the C_{60} layer and they could act as donor sites in the film.

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