Extended Abstracts of the 2011 International Conference on Solid State Devices and Materials, Nagoya, 2011, pp690-691

Direct Observation of Charge Carrier Concentrations in Operating Field-Effect Transistors of Pentacene by Electron Spin Resonance

Hisaaki Tanaka[1], Masataka Hirate[1], Shun-ichiro Watanabe[1], Hiroshi Ito[1], Kazuhiro Marumoto[2,3], Taishi Takenobu[3,4], Yoshihiro Iwasa[5] and Shin-ichi Kuroda[1]

[1]Nagoya Univ.
Furo-cho, Chikusa-ku, Nagoya, Aichi 464-8603, Japan
Phone: +81-52-789-5165 E-mail: htanaka@nuap.nagoya-u.ac.jp
[2]Univ. of Tsukuba
Tsukuba, Ibaraki 305-8573, Japan
[3]PRESTO, Japan Science and Technology Agency (JST)
Kawaguchi 322-0012, Japan
[4]Waseda Univ.
3-4-1 Okubo, Tokyo 169-8555 Japan
[5]Univ. of Tokyo
Tokyo 113-8656, Japan

1. Introduction

Organic field-effect transistors (OFETs) have been attracting considerable attention both from scientific and technological points of view. Direct observation of charge carriers in these devices is an essential subject to understand the basic physical processes in the device operation as well as to determine the origin of the charge carriers. Field-induced ESR (FI-ESR) technique is a particularly suitable method for this purpose, providing microscopic information concerning the wave function of charge carriers and the molecular orientation at the device interface as demonstrated in regioregular poly(3-hexylthiophene) (RR-P3HT) [1] or pentacene [2] FETs.

Determination of carrier concentrations in operating OFETs is also a key issue to characterize the device properties of the organic FETs. According to the standard theory, change in the carrier concentration is induced in the FET channel by the application of drain-source voltage (V_{ds}) together with the gate-source voltage (V_{gs}), dominating the FET output current of the device. Such change has been directly demonstrated by our previous FI-ESR measurements for the RR-P3HT FETs [3]. In this study, we demonstrate the direct determination of charge carrier concentrations in operating FETs of pentacene (inset of Fig. 1), which is one of the most promising materials for the OFET, by using FI-ESR technique.

2. Experimental

The FET devices were fabricated on n^+-Si substrates with SiO_2 gate insulators [3,4]. We adopted top-contact geometry, where pentacene was vapor-deposited on 300-nm-thick SiO_2 surface treated with hexamethyldisilazane (HMDS). Au electrodes were vapor-deposited on the film. ESR measurements were performed by using a Bruker E-500 spectrometer. The spin concentration was determined from the twice-integration of the first-derivative ESR signal calibrated by that of $CuSO_4$•$5H_2O$. All the measurements were carried out at room temperature. The typical mobility of the present devices was obtained as μ = 0.1 cm^2/Vs with a threshold voltage of V_{th} = -20 V.

3. Experimental Results

Fig. 1 shows the first-derivative FI-ESR signal obtained by applying negative V_{gs} without applying V_{ds}. The external magnetic field (H) is parallel to the substrate. The FI-ESR signal is observed around g = 2.00245, which linearly increases under the application of negative V_{gs}, with a threshold voltage of -20 V, owing to the accumulation of pentacene π-electron spins at the device interface. Induced spin concentration agrees well with the charge concentration determined from the capacitance measurements, indicating that all the carriers have spin, that is, polarons.

The FI-ESR linewidth tends to become narrower as $|V_{gs}|$ increases. This indicates the occurrence of motional narrowing due to the thermal motion of carriers, which is promoted for higher voltages. This effect has been previously reported and interpreted by using multiple trap and

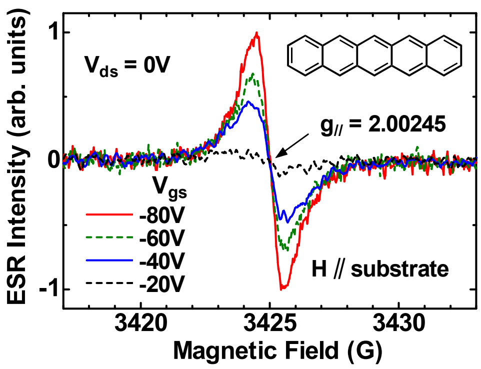

Fig. 1 First-derivative FI-ESR signals obtained with H parallel to the substrate. Inset shows chemical structure of pentacene.

release (MTR) model [5]. In this model, the trap sites tend to be filled as the charge accumulates, giving higher carrier motion for higher carrier concentration. We note that the higher FET mobility for higher $|V_{gs}|$ is also observed in the present study, showing a phenomenological relation of $\mu \propto V_{gs}^{\beta-2}$ with $\beta = 2.6$.

When the drain voltage is applied together with the gate voltage, concentration of the carriers (N) exhibits distinct V_{ds} dependence as shown in Fig. 2(a). To obtain the accurate value of N, the contribution of the carriers accumulated underneath the source and drain electrodes of the top-contact device are subtracted from the total carrier concentration. Obtained carrier concentration within the channel, normalized by the value at $V_{ds}= 0$, decreases gradually in the linear region, reaching to ~ 70 % from the initial value at the pinch-off voltage ($V_p=V_{gs}-V_{th}$), whereas it becomes almost V_{ds}-independent for the saturation region.

According to the gradual channel approximation, local carrier concentration $Q(x)$ within the channel reflects the local potential $V(x)$ through the relation $Q(x) = C_i\{V(x) - (V_{gs} - V_{th})\}$, where C_i denotes the capacitance of the gate insulator. If we assume a linear potential distribution, the total carrier concentration becomes 50 % of the initial value at $V_{ds} = V_p$ [3]. However, the carrier concentration reaches clearly higher value of ~ 70 % at $V_{ds} = V_p$ in the present device. This result indicates that the potential changes moderately toward the drain electrode in the channel region by applying V_{ds}, whereas it exhibits steep drop near the drain electrode, as schematically shown in Fig. 2(b) by adopting the bottom-contact geometry for simplicity. At the pinch-off voltage, the conductance at the drain electrode is very small due to the carrier depletion, and hence, the lateral electric field increases steeply at the drain electrode in order to satisfy the current continuity condition. This is the origin of the potential drop near the drain electrode. Such

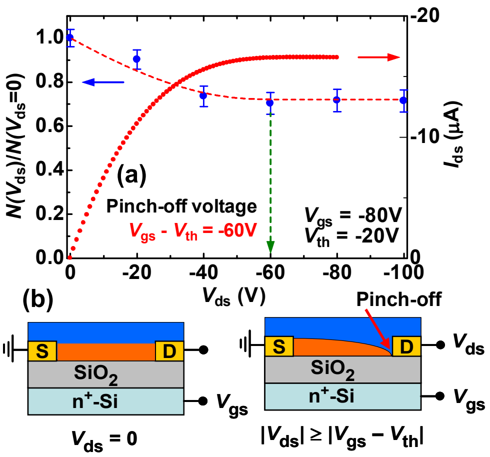

Fig. 2 (a) V_{ds} dependence of the carrier concentration (N) normalized by the value at $V_{gs} = 0$ together with the FET output current obtained for the same device. The gate voltage is -80 V. Dashed curve represents the theoretical expectation using the potential distribution with $\beta = 2.6$. (b) Schematic illustration of charge carrier distribution within the channel.

nonlinear potential profile has been reported by using Kelvin probe force microscopy (KPFM) measurements [6,7]. In addition, if the mobility depends on the carrier concentration, potential drop around the drain electrode becomes more significant due to the large change of the conductance. By taking these effects into consideration, obtained V_{ds} dependence of the carrier concentration is compared with that expected from the theoretical potential distribution in the case of $\beta = 2.6$ [7] as shown by the dashed curve in Fig. 2(a). The agreement between the observed and calculated results is satisfactory.

We also note that the pinch-off voltage of $V_p = -60$ V in the present device is reasonably determined from the V_{ds} dependence of the carrier concentration as shown by the dashed arrow in Fig. 2(a), which is consistent with the constant output current (I_{ds}) for $V_{ds} < -60$ V in the saturation region of the same device.

4. Conclusions

In this work, we demonstrated the change of charge carrier concentration in the operating FET of pentacene through the accurate determination of spin concentration by the FI-ESR technique. The carrier concentration decreased to ~70 % at the pinch-off point of $V_{ds} = V_{gs} - V_{th}$ from the initial value at $V_{ds} = 0$ and stayed nearly constant in the saturation region. The results agreed well with the potential distribution reported by using KPFM measurements. The gate voltage dependence of the carrier mobility was microscopically confirmed from the motional narrowing of the FI-ESR signal, which also affects the carrier concentration in the operating devices.

Acknowledgements

This work has been partially supported by Grant-in-Aid for Scientific Research (Grants No. 22340080) from the Ministry of Education Culture, Sports, Science and Technology of Japan.

References

[1] K. Marumoto et al., J. Phys. Soc. Jpn. **74** (2005) 3066.
[2] K. Marumoto et al., Phys. Rev. Lett. **97** (2006) 256603.
[3] H. Tanaka et al., Appl. Phys. Lett. **94** (2009) 103308.
[4] S. Watanabe et al., Appl. Phys. Lett. **96** (2010) 173302.
[5] H. Matsui et al., Phys. Rev. Lett. **100** (2008) 126601.
[6] L. Burgi et al., Appl. Phys. Lett. **80** (2002) 2913.
[7] E. C. P. Smits et al., Phys. Rev. B. **76** (2007) 125202.