Photo-Induced Magnetic Field Effects in Single Crystalline Tetracene Field-Effect Transistors

Hyuk-Jae Jang[a,b,] Emily G. Bittle[b,] David J. Gundlach[b,] and Curt A. Richter[b]

[a] *Theiss Research, La Jolla, CA 92037, USA, hjaejang@gmail.com,* [b] *Engineering Physics Division, National Institute of Standards and Technology, 100 Bureau Drive, Gaithersburg, MD 20899, USA.*

Development of organic field-effect transistors (OFETs) is considered to be one of the most prominent achievements in advancing organic semiconductor device technology [1], [2]. Several decades of research on OFETs has led to significant advances in the fundamental understanding of charge carrier transport mechanisms in small-molecule organic semiconductors as well as conjugated polymers [1], [2]. However, relative to other organic-based devices such as organic light emitting diodes (OLEDs) [3] and organic photovoltaics (OPVs) [4], OFETs have not been significantly exploited to understand spin related phenomena and their associated magnetic field effects (MFEs) in organic materials even though OFETs can provide a unique scientific platform to study the dynamics of spin interactions such as the spin dependent recombination of excitons and charge transfer pairs [5], [6].

We present an investigation of the change of photo-induced current in single crystalline tetracene field effect transistors as a function of an external magnetic field to elucidate dynamics and interactions of photo-excited spin states in tetracene. Our field effect transistors consist of a heavily doped silicon substrate (gate electrode, n-type 10−3 Ω cm), 200 nm thick thermally grown silicon oxide (SiO_X) layer (gate dielectric), photolithographically defined metal electrodes made of 40 nm thick platinum on 2 nm thick titanium (source and drain contacts), and a tetracene single crystal. The tetracene single crystals were grown by physical vapor transport in a tube oven under argon flow and carefully laminated to the surface of a substrate with electrodes. Completed transistors have channel lengths of 5 μm, 10 μm, and 20 μm. A self-assembled monolayer of octadecyltrichlorosilane (OTS) was used to improve the semiconductor adhesion and to create a hydrophobic surface on the SiO_X to eliminate water [7]. Electrical measurements were carried out in a commercially available probe station at room temperature. The measurements were performed in a nitrogen gas filled environment to minimize possible device degradation by oxidation.

Fig. 1 shows the measured drain-source current (I_{DS}) *versus* drain-source voltage (V_{DS}) with different values of the gate voltage (V_G) of a single crystalline tetracene transistor with a channel length of 10 μm under an illumination intensity of 0.34 kW/m^2. The I_{DS} was much larger under illumination than in darkness (more than twice in magnitude). This increase indicates the existence of photo-induced current in tetracene. When an external in-plane magnetic field was applied, the magnitude of I_{DS} further changed. A decrease in the I_{DS} was observed (up to ≈ 4%) at an applied field of 200 mT under an illumination intensity of 0.34 kW/m^2. However, in darkness, no change in the I_{DS} was detected with an external magnetic field implying that the magnetic field dependence in tetracene is related to photo-induced phenomena. Fig. 2 displays the measured magnetoconductance (MC) at 200 mT as a function of the illumination intensity where MC is defined as $[I_{DS}(B) – I_{DS} (B = 0)] / I_{DS} (B = 0)$. We found that the magnitude of MC increased as the illumination intensity increased. In contrast, a decrease in the magnitude of MC was observed when either the V_G or the V_{DS} was increased. In addition, we found that the magnetic field dependence of MC changes when the angle between the magnetic field axis and the direction of I_{DS} was altered. Theoretical analysis suggests that this angular dependence likely resulted from the change of crystallographic orientation in the magnetic field

Tetracene is known to produce singlet fission processes under illumination, and previous luminescence and fluorescence spectroscopy studies suggested that an external magnetic field disturbs singlet fission as well as triplet fusion processes that also exist in tetracene single crystals [8], [9]. Our study on the MC of illuminated single crystalline tetracene transistors are in agreement with these previous findings. Our analysis suggests that triplet exciton-charge carrier interactions and magnetic dipolar interactions in triplets play an important role in the observed changes in I_{DS} under an external magnetic field with different illumination intensities, V_{DS}, V_G, and field angles.

References

[1] M. Muccini, “A bright future for organic field-effect transistors,” *Nature Mater*. vol. 5, pp. 605-613, 2006.

[2] H. Sirringhaus, “25th anniversary article: organic field-effect transistors: the path beyond amorphous silicon,” *Adv. Mater*. vol. 26, pp. 1319-1335, 2014.

[3] H.-J. Jang, S. J. Pookpanratana, A. N. Brigeman, R. J. Kline, J. I. Basham, D. J. Gundlach, C. A. Hacker, O. A. Kirillov, O. D. Jurchescu, and C. A. Richter, “Interface engineering to control magnetic field effects of organic-based devices by using a molecular self-assembled monolayer,” *ACS Nano* vol. 8, pp. 7192-7201, 2014.

[4] B. Hu, L. Yan, and M. Shao, “Magnetic-field effects in organic semiconducting materials and devices,” *Adv. Mater.* vol. 21, pp. 1500-1516, 2009.

[5] M. Nishioka, Y.-B. Lee, A. M. Goldman, Y. Xia, and C. D. Frisbie, “Negative magnetoresistance of organic field effect transistors,” *Appl. Phys. Lett*. vol. 91, pp. 092117-092119, 2007.

[6] T. P. I. Saragi and T. Reichert, “Magnetic-field effects in illuminated tetracene field-effect transistors,” *Appl. Phys. Lett*. vol. 100, pp. 073304-073306, 2012.

[7] E. G. Bittle, J. I. Basham, T. N. Jackson, O. D. Jurchescu, and D. J. Gundlach, “Mobility overestimation due to gated contacts in organic field-effect transistors,” *Nature Commun*. vol. 7, pp. 10908-10914, 2006.

[8] J. Kalinowski and J. Godlewski, “Magnetic-field effects on recombination radiation in tetracene crystal,” *Chem. Phys. Lett*. vol. 36, pp. 345-348, 1975.

[9] J. J. Burdett and C. J. Bardeen, “Quantum beats in crystalline tetracene delayed fluorescence due to triplet pair coherences produced by direct singlet fission,” *J. Am. Chem. Soc.* vol. 134, pp. 8597-8607, 2012.

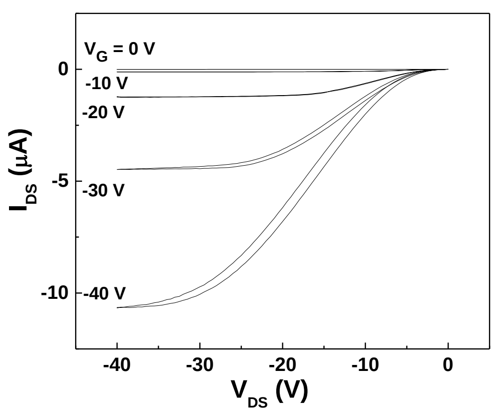

Fig. 1. Measured drain-source current (I_{DS}) *versus* drain-source voltage (V_{DS}) with different gate voltages (V_G) of single crystalline tetracene field-effect transistor under the illumination intensity of 0.34 kW/m^2 at room temperature. Gap between two electrodes was 10 µm.

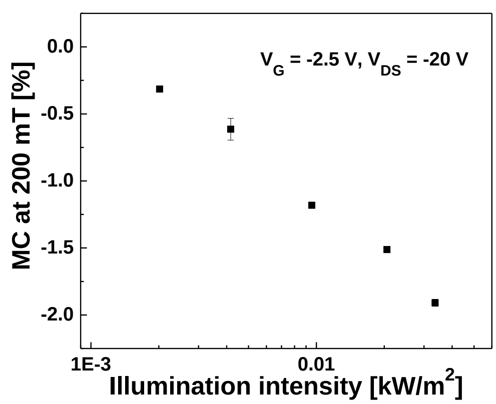

Fig. 2. Measured magnetoconductance (MC) at an external magnetic field of 200 mT *versus* illumination intensity of single crystalline tetracene field-effect transistor at room temperature. Gap between two electrodes was 10 µm.