Effect of Self-Assembled Monolayers on Sign Reversal of Organic Magnetoresistance

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Magnetic field effects (MFEs) in organic semiconductors have been of great interest to many research groups around the world since they provide a non-contact approach to control electronic and optoelectronic properties of organic-based devices by using a magnetic field [1], [2]. The MFE on electrical resistance of non-magnetic organic semiconductors, organic magnetoresistance, shows promise for use in novel organic spintronic devices. While a variety fundamental models have been proposed to explain the effect, there is no consensus on the physical mechanism (or mechanisms) causing organic magnetoresistance [3]. It has been found that the magnitude and even the sign of organic magnetoresistance can change by varying measurement and fabrication conditions such as bias voltage, temperature, and film thickness, whose origin is suggested to be a competition between different MFE mechanisms [4], [5].

In this work, we demonstrate the ability to manipulate organic magnetoresistance in Alq_3 (tris-(8-hydroxyquinoline) aluminum) – based devices by simply adding a self-assembled monolayer (SAM) at the surface of the electrodes. SAMs have been known for their versatile use in various technological applications including electronics, micro- and nano-mechanical systems, electrochemistry, and even bio-technology [6]. Among their many interesting characteristics, it has been found that SAMs can alter the physical property of the surface of metals and thus modify the interface between a metal electrode and organic thin film when they are inserted [7]. We report for the first time on how the interfacial modification by using SAMs can change the sign of organic magnetoresistance in organic-based devices.

Fig. 1 schematically illustrates our device structure. Fig. 2 displays electrically measured organic magnetoresistance of two different Alq_3 devices, one with fluorinated SAM (heptadecafluoro-1-decanethiol $[CF_3(CF_2)_7(CH_2)_2SH]$ or F-SAM) between the Au electrode and the Alq_3 and one without it; the latter shows a negative magnetoresistance but, the former displays a positive magnetoresistance despite having the same current density at room temperature.

In order to understand charge carrier transport and its accumulation in our Alq_3-based devices and thus elucidate the root cause of the change in the organic magnetoresistance of these devices, we carried out impedance spectroscopy measurements and found that the device with F-SAM induces the accumulation of charge carrier at the Au electrode interface in contrast to the one without F-SAM. To obtain a complete picture of interfacial, topological, and crystalline properties of our devices, we performed UPS (Ultraviolet Photoelectron Spectroscopy), XPS (X-ray photoelectron spectroscopy), XRD (X-ray diffraction), and AFM (atomic force microscopy). Further experiments revealed that the inclusion of a thin TPD (N,N′-Bis(3-methylphenyl)-N,N′-diphenylbenzidine) layer along with F-SAM insertion creates double charge accumulation barriers which enable the manipulation of the organic magnetoresistance by an external bias voltage.

References

[1] B. Hu and Y. Wu, "Tuning magnetoresistance between positive and negative values in organic semiconductors," *Nature Mater*. vol. 6, pp. 985-991, 2007.

[2] J. Kalinowski, M. Cocchi, D. Virgili, P. Di Marco, and V. Fattori, "Magnetic field effects on emission and current in Alq3-based electroluminescent diodes," *Chem. Phys. Lett.* vol. 380, pp. 710-715, 2003.

[3] W. Wagemans, P. Janssen, A. J. Schellekens, F. L. Bloom, P. A. Bobbert, and B. Koopmans, "The many faces of organic magnetoresistance,". *SPIN* vol. 1, pp. 93-108, 2011.

[4] J. D. Bergeson, V. N. Prigodin, D. M. Lincoln, and A. J. Epstein, "Inversion of magnetoresistance in organic semiconductors," *Phys. Rev. Lett.* vol. 100, pp. 067201-067204, 2008.

[5] P. Desai, P. Shakya, T. Kreouzis, and W. P. Gillin, "The role of magnetic fields on the transport and efficiency of aluminum tris(8-hydroxyquinoline) based organic light emitting diodes," *J. Appl. Phys*. vol. 102, pp. 073710-073714, 2007.

[6] J. C. Love, L. A. Estroff, J. K. Kriebel, R. G. Nuzzo, and G. M. Whitesides, "Self-assembled monolayers of thiolates on metals as a form of nanotechnology," *Chem. Rev*. vol. 105, pp. 1103-1169, 2005.

[7] W. H. Lee, J. H. Cho, and K. Cho, "Control of mesoscale and nanoscale ordering of organic semiconductors at the gate dielectric/semiconductor interface for organic transistors," *J. Mater. Chem*. vol. 20, pp. 2549-2561, 2010.
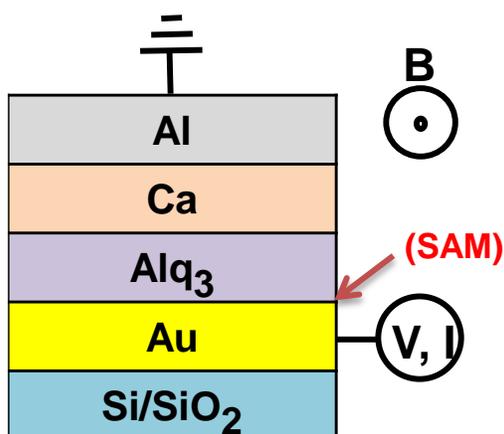
Fig. 1. Schematic view of Au (100 nm)/Alq₃ (100 nm)/Ca (20 nm) device and the illustration of electrical measurement and magnetic field set-up.
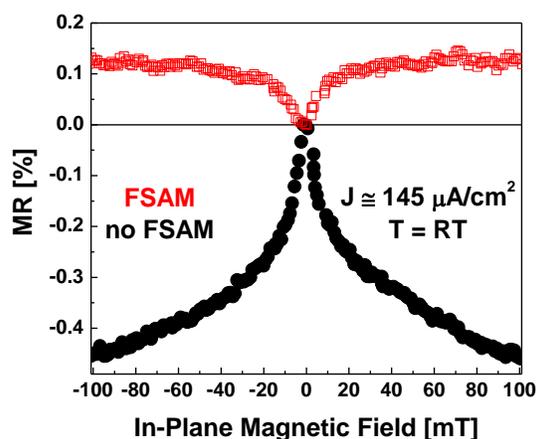
Fig. 2. Magnetoresistance of two different devices; Au/Alq₃/Ca (black filled circles) and Au/F-SAM/Alq₃/Ca (red open squares) at room temperature