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Effect of Space-Charge Field on Injection Properties in Organic Electronics

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1. Introduction

Nanoparticle researches in last decades found various applications in electronics. Several papers have recently reported on memory devices or single-electron devices [1-2]. These effects are based on Coulomb blockade, existence of energy well with barrier higher than thermal energy and charge transport phenomena in nanoscale materials [3]. Their capabilities dwell in exceptional potential in accumulation of charge. These properties make NPs promising candidates for future nanoscaled architecture to fabricate microelectronic devices. On other hand, recently used organic field-effect transistor (OFET) devices based on organic semiconductors exhibit low mobility and carrier transport is mostly limited by space-charge effect. It has been shown that carriers injected from a Source electrode dominate OFET operation. Therefore, the charge accumulated on semiconductor-gate insulator interface has great influence on device properties.

In this contribution, we will report on behaviour of metal/pentacene/metal (MIM) structure with gold-aluminium electrodes. By including a single layer of silicon nanoparticles as trapping centers, it was possible to create a space-charge field with strong influence on carrier injection. Modification of injection properties was carefully tested also on OFET with golden electrodes. All characteristics are compared with the reference sample (without NPs) prepared in the same way. We found that NPs work as trapping centers and can serve for design of accumulated charge in OFET. Thus adjustment of injection and transport properties is possible. This behaviour is fully supported by theoretical analysis of the Maxwell-Wagner model**.**

2. Experimental results and Discussion

Samples used in the experiments were MIM structures and top-contact pentacene OFETs. Glass substrates and heavily-doped Si wafers with a thermally prepared silicon dioxide (SiO_2) insulating layer were used as the base substrates for MIM and OFET, respectively. Silicon NPs stabilized with sodium n-dodecylbenzenesulfonic acid (DBSA) and with size of ~5 nm were spin-coated before the pentacene deposition. On the NPs film, approximately 2-3 layers thick (~25 nm), pentacene (Pen) was deposited by thermal evaporation and subsequently gold (Au) or aluminium (Al) electrodes, see inset in Fig.1. For comparison, in the same way the reference sample (without NPs) was prepared as well as various NP concentrations.

Created structures were investigated by standard current – voltage analysis, as well as impedance spectroscopy (IS). For OFET, optical second harmonic generation (SHG) technique, which is a powerful tool for injection property and mobility determination, was also applied.

Prepared Al/Pen/Au MIM structure exhibits well-known blocking contact for Al electrode, see Fig. 1. In the analysis of impedance spectroscopy measurement, the Schottky injection behaviour and total blocking on metal-organic interface on Au and Al was observed, respectively. This is in full accordance with energy barrier estimated from difference between the Fermi energy of the metal (5.1 and 4.3 eV for Au and Al, respectively) and highest occupied molecular orbital (HOMO) of pentacene with energy of 5.1 eV.

On the other hand, although slight reduction of number of carriers injected from golden electrode was noticeable with the inclusion of the silicon NPs layer, a strong increase of current flow from the aluminum electrode was observed (Fig. 1). Similar behaviour was recorded by IS (data not shown here). By IS, we were able to distinguish the interface and bulk effects related to injection and transport properties, respectively. Both electrodes (Au, Al) show Schottky injection. Surprisingly, the symmetric behavior of carrier injection points out that there is strong decrease of Al-electrode hole injection barrier.

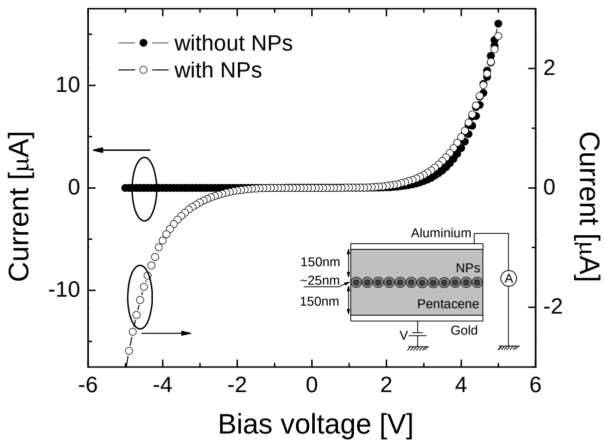

Fig. 1 Dependence of current on bias voltage for MIM structure without or with NP layer as a space-charge . Positive and negative bias voltage corresponds to holes injection from gold and aluminium electrode, respectively. Inset shows analyzed MIM structure.

The following hypothesis was formed to explain this behavior. Charge carriers injected from the gold electrode are partially trapped by the NP layer; thus a sheet of space-charge is created in the device. The created electric field consequently decreases the energy barrier for carrier injection from the Al electrode.

To verify this assumption, experiments with lower density of NPs were performed. It was shown that by slight decrease of NP density we are able to receive transition between limit cases presented in Fig. 1.

Moreover, for detail analysis of the space-charge effect on injection and transport properties, OFET structure with NP layer on the pentacene-SiO_2 interface was studied. Few different effects were observed. First, the trapped charge has a strong influence on threshold voltage shift. This shift can be easily modified by various densities on NPs. Second, in the presence of NP layer, a change of transport properties was observed. This change was successfully analyzed on the basis of the Maxwell-Wagner effect model [4] as an absence of accumulated charge layer on semiconductor-gate insulator interface. Output characteristics in the saturated region indicate mobilities for OFET without and with nanoparticles are 1.9×10^{-2} $cm^2/V.s$ and 0.8×10^{-3} $cm^2/V.s$ respectively.

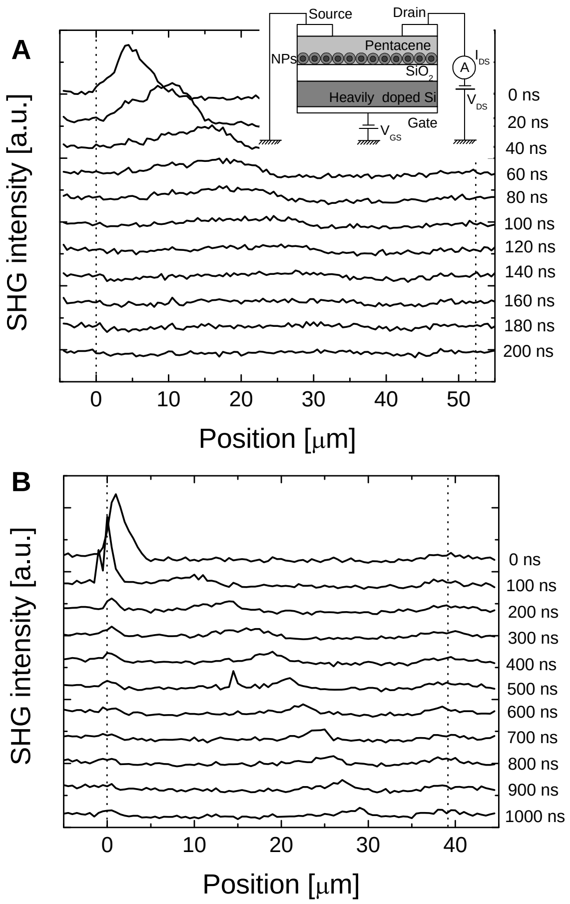

Fig. 2 TRM-SHG measurement of OFET (A) without NPs and (B) with NPs. Inset shows the analyzed OFET structure.

For accurate estimation of injection properties, measurement of time-resolved microscopy SHG (TRM-SHG) was done. (Fig. 2) This novel technique [5] enables us to record electric field evolution in OFET after application of boxcar pulse of 100V. Thus carrier injection and transport properties represented by potential drop and bulk mobility can be estimated. The potential drop on the electrode is much higher in case of NPs sample (Fig. 2b), represented by remaining SHG peak at the edge. Peak movement, which represents the mobility in the OFET channel, is also markedly slower when NPs are present.

Our analysis based on [6] of pentacene OFET showed mobility of 0.39 $cm^2/V{\cdot}s$ and nearly zero potential drop as well as injection time. In contrast, pentacene OFET including NP layer exhibits only 0.08 $cm^2/V{\cdot}s$ and injection time of ~200ns. The discrepancy of mobility decrease due to NPs from I-V and TRM-SHG measurements (~24 and ~5 times) can be attributed to injection properties represented by contact resistance (R_c). Moreover, on the metal-organic interface remains potential drop of ~20V (related to R_c). This fully corresponds to our idea of trapped charge in the NP layer.

3. Conclusion

On the basis of presented results we can conclude that incorporation of NPs into the organic electronic devices, such MIM or OFET structure, is reasonable for set-up of carrier injection and transport properties by creation of space-charge field. Moreover, this gives us a powerful tool for control of carrier injection time and design device properties independently on electrode materials. In this sense, it is possible to avoid electric breakdown which is typical for high injection barrier electrodes. Therefore design of space-charge is crucial for next organic semiconductor applications.

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