

Local carrier dynamics in organic thin film transistors investigated by time-resolved Kelvin probe force microscopy

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ABSTRACT

Understanding the relationship between the microscopic structures and the carrier transport properties in organic electronic materials is of fundamental interest to exploit the potential of the materials and thus to realize highly efficient organic devices. In this study, carrier ejection processes in polycrystalline organic thin film transistors are investigated by time-resolved Kelvin probe force microscopy (tr-KPFM) that is capable of visualizing time evolution of the surface potential triggered by the application of a voltage pulse with a temporal resolution of 3 ms. The obtained tr-KPFM images show that the spatial distribution of the diffusing charge carriers is mostly non-homogeneous, reflecting the complex resistivity distribution in the channel region formed by the crystalline grains. The trapped charges in some grains diffuse to the electrodes on a time scale of several tens of milliseconds due to the low contact resistance of the grain boundaries, while most of the trapped charges diffuse on a time scale of hundreds of milliseconds.

1. Introduction

Carrier behavior in organic electronic materials is of fundamental interest to exploit the potential of the materials and thus to realize highly efficient organic devices such as organic light-emitting diode, organic photovoltaic cells and organic thin film transistors (OTFTs) [1–3]. It is well-known that the performance of organic thin film devices is associated with their molecular packing and crystallinity [4], and that the carrier transport properties of the thin film devices are mostly limited by the boundaries between the crystalline grains and the interface between the grains and metal electrodes [5]. The relationship between the microscopic structures and the carrier transport properties have been studied using various analytical techniques [6] as well as conventional transport measurements [7]. However, as most analytical tools do not have sufficient spatial and temporal resolutions at the same time, the microscopic nature of the carrier behavior and their dynamics in the organic thin films has not been fully elucidated.

Kelvin probe force microscopy (KPFM), a surface potential mapping technique having a nanometer-scale spatial resolution, and other similar techniques have often been used to investigate the OTFTs and other organic devices [8–19]. While they have mainly been applied for acquiring surface potential maps or profiles of the operating OTFTs to identify the current limiting regions in early studies [9–11], they have also been recently used to characterize the trapped charges in the OTFT

channel [12–18]. These techniques are not only applicable for measuring the static potential distribution, but also useful for investigating the carrier dynamics in the device. Specifically, several researchers have investigated the dynamics of the trapped charges in the OTFT channel by recording the surface potential variation as a function of the gate voltage that switches the device on and off [13,16]. We recently demonstrated visualization of the trapped charges being ejected from the OTFT channel using KPFM with a gate-voltage sweep [18]. However, since the technique is based on the recording of the surface potential versus gate voltage curve at each pixel, it only provides a snapshot of the redistribution process of the trapped charge at a time scale of the gate voltage sweep time, which is typically on the order of hundreds of milliseconds.

Here, we demonstrate visualization of the decay process of the trapped charges in the OTFT channel using time-resolved KPFM (tr-KPFM) [20] with a temporal resolution of 3 ms, which is two orders of magnitude faster than the previous method [18] while keeping a high spatial resolution on the order of tens of nanometers. The tempo-spatial distribution of the trapped charges gives a two-dimensional map of the trapped charge carrier lifetime, which reflects the microstructures of the thin film in the escape pathways towards the electrodes. We also demonstrate that the effective charge carrier mobility can be estimated by performing a numerical simulation of the time evolution of the potential distribution.

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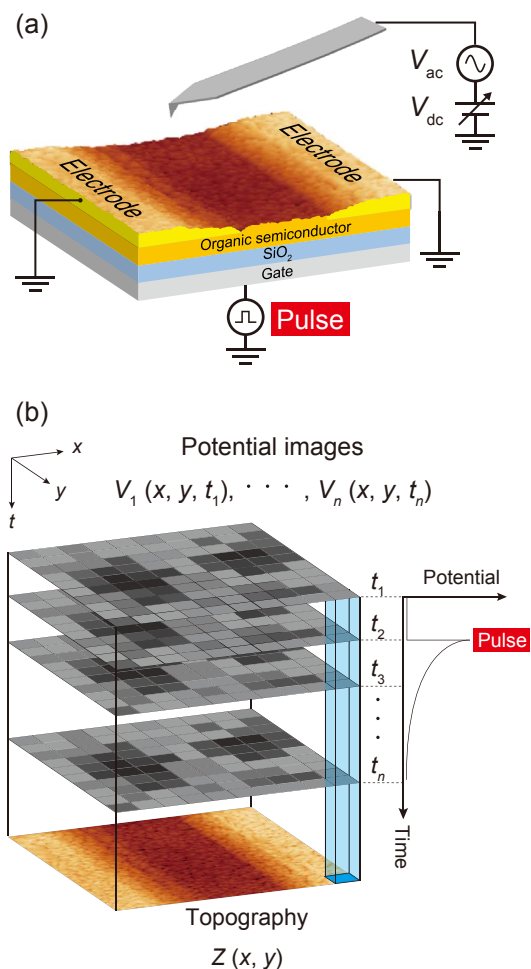


Fig. 1. Schematic illustrations of the tr-KPFM measurement performed on a DNTT-OTFT. (a) Experimental setup of the tr-KPFM. While a tip is raster-scanned on a sample, the feedback control of a tip-sample distance and the raster scan are turned off at each pixel for a certain period of time. During this period, a voltage pulse is applied to the gate and the surface potential is recorded for n samples. (b) n frames of surface potential images can be reconstructed from the collected three-dimensional surface potential data.

Fig. 1(a) shows a schematic diagram of the tr-KPFM measurements. For KPFM, an ac modulation voltage (V_{ac}) and a dc voltage (V_{dc}) were applied to the tip, and the V_{dc} was controlled by a feedback circuit such that it matched the surface potential (V_{sp}). The time evolution of the V_{sp} triggered by applying a voltage pulse was measured at every pre-configured pixel (e.g., 64×64 matrix). A voltage pulse was applied to the gate while the source and drain electrodes were electrically grounded, and the response of the surface potential was measured for a certain period of time, typically 1 s. Before each application of the voltage pulse, the feedback control of the tip-sample distance was turned off such that it did not respond to the electrostatic force during each measurement. After acquiring the time evolution of the surface potential, the feedback control was resumed and the tip was laterally moved to the next pixel. By repeating these procedures, three-dimensional (3D) surface potential data were collected. The collected data were reconstructed into sequential surface potential images, as shown in **Fig. 1(b)**.

2. Materials and methods

We used a modified commercial AFM instrument (JSPM-5200, JEOL). Platinum-coated silicon cantilevers (OMCL-AC240TS, Olympus) with a nominal spring constant of 2 N/m and a resonance frequency of

70 kHz were employed. The tip-sample distance was regulated by the constant frequency shift mode ($\Delta f = -20$ Hz), while the cantilever was self-oscillated with an amplitude of 20 nm. A modulation voltage ($V_{ac} = 2$ Vp-p, 1 kHz) was applied to the tip and the modulation of the frequency shift was detected by a home-built phase-locked loop circuit and a lock-in amplifier (HF2LI, Zurich Instruments). A bias voltage generated by the KPFM feedback circuit, which compensates the contact potential difference between the tip and the sample, was also applied to the tip and recorded as V_{sp} . All the tr-KPFM measurements were performed in a vacuum environment ($< 8 \times 10^{-3}$ Pa).

We fabricated an OTFT with a bottom-gate top-contact configuration using a dinaphtho[2,3-*b*:2',3'-*f*]thieno[3,2-*b*]thiophene (DNTT) thin film. The devices were fabricated on a heavily-doped n-type Si wafer with a thermally-grown oxide layer of 300 nm. The Si substrate was rinsed with ethanol and treated by UV ozone cleaner for 30 min prior to the device fabrication. To passivate the surface of the oxide layer with self-assembled monolayers, the substrates were enclosed in a Teflon vessel with octadecyltriethoxysilane and placed in an N_2 atmosphere oven at 150 °C for 2 h. After the treatment, the substrates were washed with toluene, acetone, ethanol, and deionized water. A 60-nm-thick DNTT film and a 40-nm-thick gold were successively deposited by vacuum evaporation. During the deposition of the gold, a carbon fiber with the diameter of 5 μ m (IMS65, Toho Tenax) was used in combination with an aluminum shadow mask to define the channel region. The channel length of the fabricated OTFT was about 3.5 μ m.

3. Results

3.1. tr-KPFM measurements of OTFTs

A typical time evolution of the surface potential recorded on the center of the OTFT channel is shown in **Fig. 2**. The surface potential was measured for 1 s. The device was turned on at $t = 20$ ms and turned off at $t = 50$ ms by applying the voltage pulse (-2.5 V, 30 ms) to the gate electrode. The surface potential in the channel region was about 240 mV before applying the gate pulse. When the negative gate bias was applied to the gate while the source and drain were electrically grounded, positive charge carriers were immediately injected from the top electrodes to the channel region. This resulted in the molecular energy level shift of the DNTT layer with respect to the Fermi level [21] and the surface potential immediately decreased to about -260 mV. On the other hand, after the gate bias was turned off, the surface potential once increased to about 2 V and slowly decreased to the initial surface potential with a decay constant of greater than 1 s. The observed decay in the surface potential was attributed to the positive

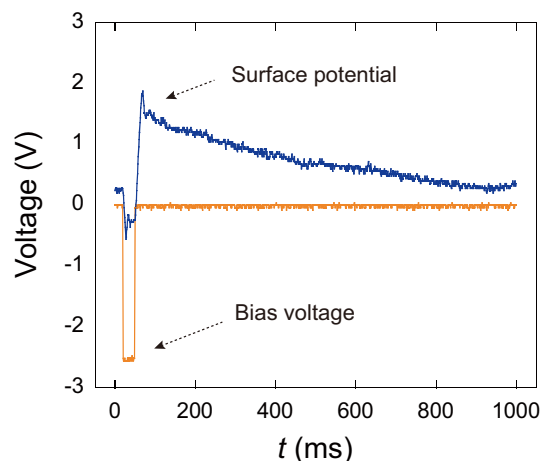


Fig. 2. Typical time evolution of the surface potential recorded in the center of the channel region when the voltage pulse of -2.5 V with a width of 30 ms was applied to the gate electrode at $t = 20$ ms.

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