

論文の内容の要旨

論文題目 Study of Carrier Trapping Effects in Pentacene Thin Film Transistors
(ペンタセン薄膜トランジスタにおけるキャリアトラッピング効果に関する研究)

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Electronic devices based on organic semiconducting crystals, such as thin film transistors (TFTs) and light emitting diodes (LEDs), have attracted considerable interest as possible inexpensive and flexible alternatives to inorganic devices. As one of the most prominent members of organic semiconductor applications, pentacene exhibits a carrier mobility of up to about $3 \text{ cm}^2/\text{V}\cdot\text{s}$. However, despite the enormous progress in performance improvements, these organic devices show the crucial instability of a threshold voltage shift (ΔV_T) or hysteresis brought about by bias stress. From a practical viewpoint, this characteristic has been a major drawback for the organic device application compared to those of inorganic devices. Because the organic device instability results predominantly from the charge trapping phenomena, it shows that the systematic investigation of charge trapping effects both, spatial and energetic, is vital to this field's progress. Moreover, nowadays, ambipolar transistors using a single organic material as their active layer have become a great issue in the research of organic electronics, which can open new fields of applications for organic electronics. Although the minority carrier effect in the p-channel in organic devices has been ignored so far, this fact also reflects that an important point is to understand the contribution of both carriers (electron and hole) related to the material property and the device physics within a single active material for developing organic electronics.

This work outlines our findings in understanding and characterizing of the charge trapping features and their mechanisms in pentacene thin film transistors. The primary goal of this thesis is to contribute and enhance our understanding of the charge trapping nature in organic electronic devices. Here our study of the charge trapping effect related to bias-induced device instability has been followed from the systematic viewpoint of real space and energy space. In *real space*, the device instability feature was analyzed with respect to the effect of molecular structures and their ordering in the crystallized film, which addresses the *hole trapping effect* in the spatial channel region. In *energy space*, we investigated the charge trapping phenomena through spectroscopic and electrical analysis, which also show the

predominant *electron trapping effect* and its substantial role in causing device instability. The main findings and achievements in this thesis are summarized as below.

1. The comparative study of the ΔV_T behavior and thin film phase transition reveals that the hole trapping aspects are strongly influenced by the molecular structural order, particularly near the pentacene/SiO₂ interface in the channel. The growth temperature dependence of the ΔV_T feature shows that an increase in external bias stress accelerates the hole trapping, resulting in a further shift of V_T and the larger grain size of pentacene films grown at high temperature is subject to more significant hole trapping. The structural arrangement of molecules in the film is favored in a fashion similar to that of the single “thin film” phase to suppress hole trapping sites. In the film's two crystallographic phases, phase mixing near the interfacial region might contribute to hole trapping sites due to the structural disorder in different crystallographic orientations. Moreover, we show the effect of the chemically-modified surface of SiO₂ on reducing the device's ΔV_T . A functionalized surface with alkyl chain functional groups suppresses the trapping sites at the interface and in the channel adjacent to the dielectric surface by reducing the mismatched interface state and by yielding a highly ordered molecular structure. This result reveals that hole trapping can be effectively suppressed by controlling the surface free energy to a value close to that of the pentacene layer.

2. We found that the device instability increases significantly when it is exposed to photo-irradiation with a reverse bias. This fact reveals that a number of electron trapping sites are located in the energy level of the pentacene band gap. Clear evidence was presented for the existence of electron trapping sites at the pentacene/SiO₂ interface. The mechanism of the photoinduced threshold voltage shift was presented by providing a ΔV_T model governed by electron trapping. In addition, we observed that the trapped electrons' lifetime is well described by two time constants, which are thought to be associated with different energy trapping levels in the forbidden gap state of pentacene.

3. The systematic investigation using spectroscopic analysis identifies that two kinds of electron trap states are distributed above and below the energy of 2.5 eV in band gap of the pentacene crystal. The study of the photocurrent spectra enables detailed comprehension of electron trapping aspects within the pentacene band gap and their trapping level in energy space. The shallow and deep trapping states are distributed at two centroidal energy levels of 1.8 and 2.67 eV in the pentacene band gap. Moreover, we suggest possible reasons for the shallow trap state with some suggestions for suppressing the trapping sites. The comparative study of trapped charges and film properties through thermally stimulated current (TSC) characterization shows that the shallow state of pentacene crystal is also composed of two kinds of traps, strongly associated with the film quality, which would originate from SiOH silanol groups on the dielectric surface and the structural defects in the crystallized pentacene film. Finally, we

propose the systematic energy profile of electron trap states in the pentacene crystal for the first time.

We believe that these results help to understand charge trapping effects related to the material properties and the device physics, and help to realize ambipolar transistors using pentacene material. This work also will provide a good framework and a new direction for progress in this field in the future.