博士論文要約（Summary）

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論文題名（外国語の場合は、その和訳を併記） Thesis Title（foreign language title must be accompanied by Japanese translation）
Basic Studies for THz Imaging Sensors Using Organic Field Effect Transistors 有機電界効果トランジスタを利用した THz イメージングセンサの研究

As the last unexploited region of the electromagnetic spectrum, terahertz (THz) wave, of which frequency is ranging from 0.1 to 10 THz, has been attracting a great deal of attention and the growth of research into THz waves is driven by scientific and industrial applications in areas such as defense, security, biology, and medicine. Remarkable progress of its applications has been made in recent years, however, manufacturing innovative flexible large-area THz sensing devices with high sensitivity and low cost has been an important challenge to be undertaken. In this thesis, I propose applying the HOMO-band edge fluctuations (about 10 meVrms) that reproducibly appears in organic field effect transistors (OFETs) with pentacene active layers to the sensing element of flexible THz-imaging devices.

Grain boundaries in a polycrystalline active layer deteriorate the electrical performance of an OFET because they generate potential barriers to carrier transport. Such barriers not only reduce the apparent field-effect carrier mobility, they also determine the thermal activation energy for carrier transport and make the mobility very sensitive to the operating temperature. Potential barriers are predicted to be diminished once the oriented growth of grains is achieved. However, organic thin films grown on amorphous surfaces such as SiO_2 or polymers inevitably consist of randomly oriented polycrystalline grains.

Several studies have found that pentacene, a typical organic material for OFETs, or α-sexithiopene tends to preferentially orient against sharp slope edges on substrates by graphoepitaxy. Graphoepitaxy is a phenomenon in which the macroscopic profile of the substrate surface restricts the orientation of growing crystals through the minimization of the free energy of crystals on the surface. It is a promising technique for achieving oriented growth of crystalline films on amorphous substrates. If an array of such slope edges on the entire substrate surface causes all the grains to align in a single direction, then there would be fewer grain boundaries with large potential barriers and consequently the field-effect carrier mobility is expected to increase.

In this thesis, graphoepitaxy method was employed to improve the carrier transport properties of OFETs and to increase the sensitivity of the THz sensors. By controlling the in-plane orientation of pentacene crystals using periodic grooves with slope edges, density of polycrystalline domain boundaries that induce large potential barriers in pentacene was expected to decrease. Figure 1 shows an AFM topographic image of pentacene grains in the patterned OFET (growth rate: 6.6 nm/min). From four similar images taken on different locations, we determined that about 28% of the grain boundaries have small-misfit-angles.

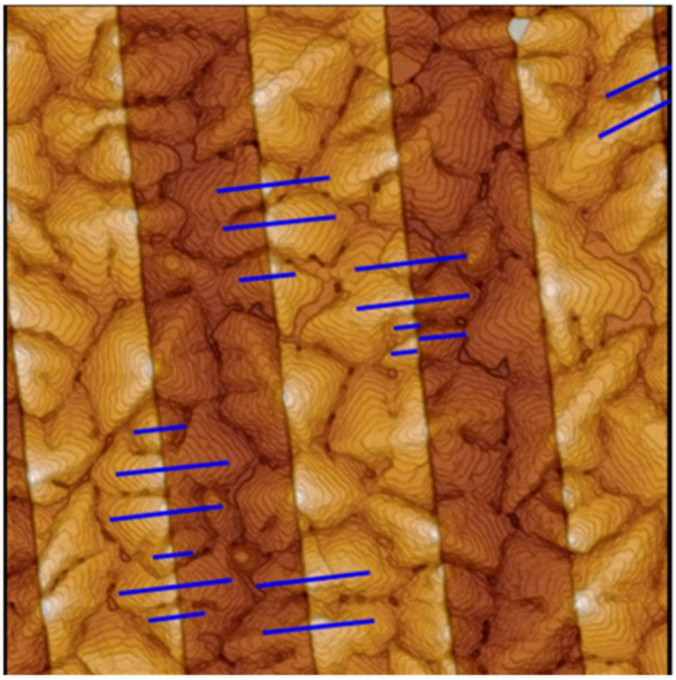

Fig.1 AFM topographic image (10×10 μm^2) of pentacene grains grown at 60°C and 6.6 nm/min on patterned substrates. A high-pass-filtered image is combined with the corresponding color-scale image to enhance the shape of the molecular steps. Blue lines indicate parallel in-plane orientation of adjacent pentacene grains.

This ratio agrees well with the expected probability from the orientation distribution. However, it is much smaller than the ratio of additional barrierless boundaries, 58%, which is estimated from the grain size dependence of the mobility. There must therefore be another factor that reduces the high-barrier boundary. Consequently, the patterned substrate is considered to affect the apparent mobility of pentacene polycrystalline films not only by the controlled orientation of the grains but also by alignment of the grains along the current direction.

We demonstrated that the in-plane orientation of pentacene grains could be controlled by periodic grooves with slope edge structures. Nearly half of the grains were aligned so that their b-axes were perpendicular to the groove edge. Compared to the flat OFETs, the patterned OFETs had a 10–20% higher field-effect mobility. This increase in the mobility is relatively small because the average grain size decreases when a patterned substrate is used and when the deposition conditions remain the same. By accounting for the influence of grain size, the average barrier height at grain boundaries in the patterned OFETs was considerably smaller (132 meV) than that in the flat ones (157 meV). The mobility could be further improved by optimizing the deposition conditions for the patterned substrate. On the other hand, the number of large-barrier grain boundaries was estimated to be reduced more than that expected based on the grain orientation distribution. By the maximum degree of orientation obtained in this work, the improvement of the sensitivity was estimated to be 30%.

As the first stage of the THz sensor mechanism, it was confirmed by THz time-domain spectroscopy (THz-TDS) that accumulated free holes in the pentacene channel could receive energy from THz photons. Pentacene OFETs were fabricated on n-type silicon substrates covered with 300-nm-thick oxide layers. The substrates were served as gate electrodes to bias against interdigital-type source/drain electrodes on the oxide layer and the absorption spectra of the OFET were measured with THz time-domain spectroscopy (THz-TDS). Counter experiments were performed using a control sample where a very thin Au layer was deposited instead of the pentacene active layer. Transmission spectra were measured under ON-state gate bias (-30 V) of OFETs and they were normalized by OFF-state (+30 V) ones of the same sample. Figure 2 shows the modulation absorption spectra of OFET and control sample measured under the same conditions.

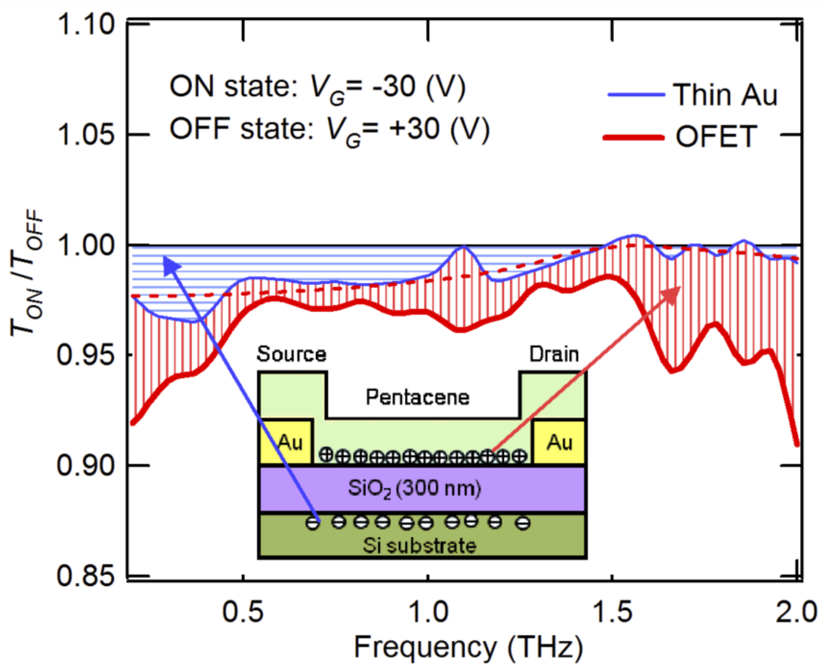

Fig.2 Modulation absorption spectra of OFET sample (red thick line) and control sample (blue thin line) obtained under the same measurement conditions. Dashed line is a theoretical curve obtained using the Drude model; it is fitted to the control sample spectrum. Horizontally and vertically hatched areas indicate absorption by the accumulated electrons in Si and by the holes in pentacene, respectively.

These two spectra have different shapes and the control sample has a lower absorption than the OFET sample. THz absorption spectra due to accumulated free holes in OFETs were obtained for the first time in the field by separating the absorption due to free electrons in the Si substrate. The THz absorption by free holes in pentacene increased when the frequency was increased from 0.2 to 2.0 THz. The Drude-Lorentz model could not account for the shape of the absorption spectra where absorption gradually increased with frequency, which suggests that the holes are weakly restricted by the potential fluctuation. The integrated absorption intensity was proportional to the transfer characteristics of the OFETs. We have found evidence that the accumulated free holes in pentacene films can be excited by THz photons to overcome the surrounding barriers in the fluctuating potential.

The results of the present work would shed new light on developing the next generation THz imaging devices with OFET structures.