和周波を用いる作動中の有機電界効果トランジスタの半導体/絶縁体界 面の研究

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Sum-Frequency Spectroscopic Study at the Semiconductor/Insulator Interface of Organic Filed Effect Transistors During Operation

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(Abstract **)** Sum-frequency generation spectroscopy (SFG) is used to study the semiconductor/insulator interface of organic field-effect transistors (OFET). Recent models of interference effects are used to deconvolute SFG spectrum of the semiconductor/insulator interface from those of all three active interfaces, for furthermore clarifying the correlation between SFG spectra and charge or hole accumulations at the semiconductor/insulator interface. The Fresnel factors of 2,7-dioctyl[1]benzothieno[3,2-b]benzothiophene on glass, gold and silicon substrates under SSP polarizations are simulated and discussed. The SFG spectra of OFET during operation under different gate voltages are obtained and discussed. The feasibility that nonresonant background and SFG intensity that can be used to monitor the accumulated charges or holes in the conducting channel quantitatively is discussed. The area of hole accumulation is restricted to a circle with radii in certain ranges. This work will help us understand and improve properties of OFET and design new organic semiconductors.

【序】 Organic field-effect transistors (OFET) become progressively interesting topics over the past decades due to widespread applications in industry [1]. The quality and reliability of the semiconductor/insulator interface plays a crucial and critical role in the ability of charge transport in the conducting channels of OFET [2]. Sum-frequency generation spectroscopy (SFG) is an inherently surface-sensitive technique to obtain vibrational spectra of molecules at surfaces and buried interface where the inversion symmetry is necessarily broken [3]. It has been utilized to study OFET since 2006, and some intriguing results about electronic properties of OFET and their SFG spectra obtained [4-6]. However, the question of interference effects has not been well dealt with in such reported works, although it becomes increasingly inevitable in the applications of SFG in recent years [7-9]. Based on the interference effects, the SFG spectra measured from OFET should be contributed from three active interfaces: the top air/semiconductor interface, the buried semiconductor/insulator interface, and the buried insulator/gate interface. It would be a motivating work to separate the SFG spectrum of the buried semiconductor/insulator interface out from the twisted SFG spectra from all three interfaces, because it is the place where charges or holes accumulate in OFET. Here we present a work to deconvolute SFG spectra of the semiconductor/insulator interface from those of three active interfaces in OFET by using recent models of interference effects. This work aims at clarifying the correlation between SFG spectra of the semiconductor/insulator interface and charge or hole accumulations in the conducting channels of OFET. It will help us understand and improve properties of OFET and design new organic semiconductors.

【方法 (実験・理論)】 An OFET sample is a top-contact structure with two gold electrodes of 30 nm thick at the top and a layer of 2,7-dioctyl[1]benzothieno[3,2-b]benzothiophene (C8-BTBT) with a thickness of 50 nm below prepared through a spin-coating method with a C8-BTBT solution in chloroform (Wako, pure grade) with a concentration of 0.8 wt%. A silicon substrate was used as a gate electrode with a SiO₂ layer of 200 nm thick coated on its surface as an insulator. There are two types of channels in an OFET sample; one is with a gap of 1.0 mm between two gold electrodes, while the other with that of 0.5 mm. An SFG spectrometer with a picosecond laser (PL2231-50) of 50 Hz repetition rate was used to study interfaces of OFET. The output energies of lasers were controlled at the minimum conditions to avoid the sample damage. The polarizations were set at SSP or PPP during measurements. The incident angles for visible and infrared lights were 65° and 55° respectively. A precision source/measure (Agilent, B2901A) was used to apply gate voltages available from 100 V to -100 V into OFET. In Fig. 1, the gate voltages were set at -60 V for the on states while 0 V for the off states.

【結果 • 考察】 Based on the models, the thicknessdependent Fresnel factors of C8-BTBT on three different substrates of glass, gold and silicon under SSP polarizations were simulated. Although they all showed thickness dependences, the Fresnel factors of C8-BTBT on silicon and gold substrates have much stronger thickness dependences than those on the glass substrate.

The SFG spectra of OFET during operation under different gate voltages were obtained. The curves of SFG intensity of I_{SFG} and nonresonant background of χ_{NR} versus gate voltages were obtained, and found to obey the equation of $I_{SFG} \propto |\chi^{(2)} + \chi^{(3)}E_0|^2$. The threshold of gate voltage for hole accumulation was inferred to be less than -3 V from the curves. Because of its high sensitivity, χ_{NR} was used to estimate hysteresis of hole



Fig. 1. Memory effects of OFET during operation of two different kinds of channels under PPP polarization

accumulation. As shown in Fig. 1, memory effects were found to exist in both two kinds of channels, but it is more severe in that with an electrode gap of 1.0 mm that that of 0.5 mm. So, $\chi_{\rm NR}$ and even $I_{\rm SFG}$ could be used to monitor accumulated charges or holes in the conducting channel quantitatively.

The position dependence on hole accumulation in the conducting channel was also explored. The area was restricted to a circle with a radius from 1.0 mm to 5.0 mm for the channel with an electrode gap of 1.0 mm, and from 0.5 mm to 5.0 mm for that of 0.5 mm.

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