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# *Investigation of organic interfaces by using optical second harmonic generation (SHG) and sum frequency generation (SFG) techniques*

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## *Abstract*

Recently, the research and the development of the organic electronic devices have received extensive attention due to their advantages such as mechanical flexibility. Understanding of the fundamental mechanism of electronic process and dynamics at surfaces or interfaces of the organic electronic devices is one of the most essential key factors to improve their performance. Many research groups already studied the electronic states of the surfaces and the interfaces of those devices by using conventional surface sensitive techniques such as photoemission spectroscopy. However, those techniques are unable to probe buried interfaces of the devices. In order to understand the electronic states and the dynamics at surfaces/interfaces of actual organic devices, a direct probing technique is required.

In this thesis, I have developed optical Second Harmonic Generation (SHG) and Sum Frequency Generation (SFG) method as a tool for monitoring the electronic and the molecular dynamics of surfaces and interfaces of organic devices directly. The SHG and SFG are second order nonlinear optical processes. These techniques are intrinsically surface/interface sensitive with sub-monolayer

sensitivity. It can be applied to the study of buried interfaces without destroying the devices. In order to demonstrate their advantage for the organic devices, I have studied the following two systems.

As the first study, I have directly observed the space charge layer (SCL) at an interface of organic layer by using SHG technique. The SCLs are known to promote electron insertion to hole transport layer from the anode or hole injection layer in Organic light-Emitting Diodes (OLEDs), although the evidence of their existence was not found directly. This study is the first investigation of the electronic states at an interface of organic devices by SHG method, and especially includes the first direct observation of the SCLs.

As the result, more peaks were observed in the SH spectra of indium tin oxide (ITO)/molybdenum trioxide ($MoO_3$)/*N-N'*–diphenyl-*N-N'*–bis(1-naphthly)-1,1'-biphenyl-4,4'-diamine (α-NPD) systems at 0.75 and 1 nm thicknesses of $MoO_3$ layer than in the spectrum of an ITO layer only. I suggest that this peak observed only for the former sample corresponds to the electronic transition at inter-hetro materials. Furthermore, I studied the change in the SH intensity from ITO/$MoO_3$/α-NPD systems as a function of thickness of $MoO_3$ layer with $2\hbar\omega$ =2.33 eV. The intensities were reduced at small thicknesses remarkably, and saturated at the thicknesses greater than 1nm. In linear spectra of the same systems, there was no remarkable dependency on the thickness, and thus the

change of the Fresnel factor of SH light was negligible. Therefore, the change in the SH intensity of the ITO/$MoO_3$/α-NPD system represents the growth and the saturation of SCL at the $MoO_3$/α-NPD interface. From the results, I demonstrated that the SHG can be used to obtain the electronic states of buried organic interfaces.

The second system is alkyl self assembled monolayers (SAMs) on quartz surfaces. Even though the use of the silane monolayers on a Si or a quartz surface is widely spread from nanotechnology to biology such as in solution-processed thin-film transistors (TFT) and protein immobilization, the packing process of the monolayers is still not well understood, since it depends on several factors such as the molecular size and functional head groups. In this thesis, I prepared alkyl silane monolayers with one and three precursors in the head group on a quartz surface, and studied the dependency of alkyl chain structure on the precursor number by using SFG method, since the key point of the formation dynamics is packing of the high flexible chains. This study is the first demonstration of the dependency of gauche defects at the alkyl chain on the precursor number of the SAMs. The gauche defects at the alkyl chains were sensitively observed by SFG spectroscopy, since the SFG is forbidden at centrosymmetric structures. In the SF spectra, the intensity of methylene vibration modes of the SAMs having one precursor group was higher than that having three. This indicates the increase of

gauche defects with decreased precursor number in the head group of the SAMs molecules.

From these studies, I have contributed to the field and industry of organic electronic devices as a technologist instructing usefulness of the SHG and SFG methods for the quality check of the products and/or finding defects in the growing processes.