# A Novel Microscope for Visualizing Electric Field in Organic Thin Film Devices Using Electric-Field-Induced Second-Harmonic Generation

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## 1. Introduction

Recently organic thin film devices have attracted much attention [1]. However understanding carrier mechnism is still insufficient for use them in electronics. The electric-field-induced optical second-harmonic generation (EFISHG) measurement is capable of visualizing carrier motions in devices, and it is very useful to analyze device characteristics [2-6]. However, the conventional optical microscope setup is not suitable for observing carrier motion in layered MIM-structure devices; At a focal plane beneath the objective lens, there is no optical electric field in the film thickness direction, but non-zero field is needed for probing carrier motion . In this paper, we report a novel EFISHG microscope that can probe electric field in the device thickness direction. The EFISHG microscope developed here has a potentiality as a tool for studying carrier motion in layerd MIM-structure devices.

# 2. Experimental

Figure 1 portrays the microscope setup developed here. Laser pulses from optical parametric oscillator (OPO), which is pumped by the third-harmonic light of Nd:YAG laser, is used as a probing light. The laser beam emitted from the OPO is homogeneous linear polarized light with a diameter of 1 cm, but after passing through a liquid-crystal polarizer [7,8], the polarization field is radial. The spatial distribution of the radial polarization field is monitored using a CCD camera equipped with a linear polarizer. As shown in Fig. 2, the radial polarization laser beam (Fig.2a) shows a bow-tie pattern after passing through a linear polarizer, while the linear polarization beam (Fig. 2b) is homogeneous after passing through it.

The resulting radial (or linear) polarized laser beam is introduced to an objective lens (100×, NA=0.9). The radial polarized laser beam that passing through the objective lens converges to generate an optical electric field in the direction normal to the surface of MIM device, whereas the linearly polarized beam resulted in a lateral optical field component only. The vertical optical field component  $E_{\omega}$ efficiently couples with a local field  $E_0$  pointing in the organic layer thickness direction. The EFISHG intensity  $I_{2\omega}$ from the device is proportional to the second-order nonlinear polarization  $P_{2\omega}$  with a doubled frequency 2 $\omega$ , given as  $I_{2\omega} \propto |P_{2\omega}|^2 = |\varepsilon_0 \chi^{(3)} E_0 E_{\omega} E_{\omega}|^2$ , (1)

where  $\varepsilon_0$  is dielectric constant of vacuum and  $\chi^{(3)}$  is the third-order nonlinear susceptibility tensor.  $\chi^{(3)}$  is a material



Figure 1 Microscope setup for measuring internal electric field in device thickness direction.

dependent parameter and enhances at a laser beam wavelength that satisfies the resonant-enhancement condition [3]. For enhancing EFISHG signal from fullerene molecules, we set the laser wavelength at 1000 nm and detected the EFISHG signal with a wavelength of 500 nm. The induced EFISHG signal is collected by the objective lens, and detected by a photomultiplier tube (PMT).

A MIM sample used for the measurement was a fullerene single-layer sandwiched between a transparent electrode and an Al back electrode. Fullerene is known as an acceptor material. We selected this material to investigate capability of the developed microscope for visualizing carrier motion in OSCs. Fullerene (thickness d=200 nm) and Al electrode were vacuum deposited on a transparent electrode, successively. The sample was set at a focal plane of the microscope. Subsequently, a DC voltage V was applied on electrode to form an electric-field (E) in the fullerene layer, E=V/d. The transparent electrode transmits probing laser beam and EFISHG signal induced. EFISHG intensity was recorded while the DC voltage was switched on and off. At last, on keeping the DC voltage at +5 V or 0V, the MIM sample was scanned at the focal plane of objective lens. In this way, the 2-dimmensional mapping of electric field in the MIM device was carried out.

## 3. Results and Discussion

Figure 2 shows EFISHG response with respect to applied



Figure 2 EFISHG response under DC voltage application. (a) Incident laser beam with radial polarization: EFISHG response is clearly seen following switching on and off voltage application. (b) Incident beam with linear polarization: EFISHG response is very small.

DC voltages, where radial or linear polarized laser beam was used to induce EFISHG signal. Results showed that axial polarized laser beam (Fig.2a) gives clear response to the applied DC voltage, whereas linearly polarized laser beam (Fig.2b) showed no response to local field formed in fullerene molecules. This result indicated radial polarized laser beam converged and formed vertical optical field in a manner as we expected. Figure 3 shows EFISHG intensity from the fullerene single-layer device, where we used radial polarized laser beam. Square-root of the EFISHG intensity, proportional to local electric field, was plotted. Linear dependence on the DC voltage suggested that the detected



Figure 3 EFISHG intensity under various voltage amplitude.

nonlinear optical signal is induced through the EFISHG process given by Eq. (1). The result showed that the developed microscope can resolve the voltage difference of 0.1 V corresponding to local electric field  $10^4$  V/cm. This demonstrates that the EFISHG microscope is capable to catch carrier motion in OSCs where 0.1 V or larger photo-voltage is generated under illumination.

Figure 4 is a 2-dimmentional electric field mapping of the fullerene sample. The EFISHG image well reflect the electric field in fullerene layer (electric field enhances blue to red colors) where +5 V is applied on electrode B and 0 V on other electrodes A, C, D.



Figure 4 (a) Optical image of the sample. (b) EFISHG image of the sample. Electric field along fullerene layer thickness direction was clearly visualized.

## 4. Conclusions

A novel microscope used for visualizing electric field in film thickness direction of layered organic device was developed. This EFISHG microscope is available to probe the electric field in device thickness direction, and it has potentiality in directly tracing carrier motion in layerd devices such as OSCs.

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