

Third Harmonic Generation of Vanadyl-Phthalocyanine Film Prepared on NaCl Substrate by Molecular Beam Epitaxy

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Abstract

The incident angle dependence of TH intensity of VOPc film irradiated with P-polarized laser beam shows an upper convex curve that has a maximum value at -15 degree. It suggests that the tensor of TH susceptibility of VOPc film is dominated with the components of $\chi^{(3)}_{xxxx}$ and $\chi^{(3)}_{xyxx}$ and also indicates that the VOPc molecules deposited on NaCl have a tilt angle. These may give us information about THG.

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1. Introduction

Organic nonlinear optical materials, which can yield a large third-order nonlinearity, have been widely studied to develop optical devices such as optically gated optical switches[1]. Organic materials with extensively delocalized π -electron systems have attracted significant attention because a big of $\chi^{(3)}$ is expected if appropriately oriented molecules can be assembled to film form. It has been demonstrated that organic materials with π -conjugated systems show remarkably large optical nonlinearities and efficient responses which are due to delocalized π -electron[2]. Conjugated linear chains such as polydiacetylenes (PDA) possess a very large third-order nonlinear optical susceptibility, as reported in both theoretical and experimental pioneering works[3]. In linear chain structures, the major contribution to the microscopic third-order optical susceptibility $\chi^{(3)}_{xxxx}$ ($-\chi^{(3)}_{xyxx}$, $\chi^{(3)}_{xyyy}$) is the main chain axis component $\chi^{(3)}_{xxxx}$ ($-\chi^{(3)}_{xyxx}$, $\chi^{(3)}_{xyyy}$) with all electric fields aligned along the chain axis (x -axis). In analogy from linear π -conjugated structures, one may take new tensor components such as $\chi^{(3)}_{xyxx}$, $\chi^{(3)}_{xyyy}$, $\chi^{(3)}_{xyzz}$, $\chi^{(3)}_{xyzz}$, $\chi^{(3)}_{xyzz}$ and $\chi^{(3)}_{xyzz}$ into consideration. Therefore, the nonlinear optical properties of the organic thin films on thermally treated substrates were investigated.

2. Experiment

The source material used was VOPc powder supplied from Eastman Kodak Company. After VOPc powder was mixed into a Kanthal-coil, it was preheated at 300°C for one hour. The substrate of NaCl was shaved just before a writing by a holder. After that, the substrate was attached on the holder. The main chamber of MBE was at about 10^{-7} Torr, and the NaCl substrate was preheated for one hour. The preheating was done at 180°C and 300°C. The evaporating temp. was kept at T_1 (evaporating temp.): 300°C. The preheating conditions of VOPc thin film were the substrate temp. (T_2 : 300°C), evaporating time (t_1 : 240 min.), annealing temp. (T_3 : 300°C) and annealing time (t_2 : 240 min.) respectively. The film thicknesses were 110nm for Sample 1 (annealing temp. of substrate: 180°C) and 120nm for Sample 2 (300°C).

3. Results and discussion

Figure 1 shows the VIS/UV spectra of Sample 1 and 2. The VIS/UV spectrum of Sample 1 has an absorption peak at 400nm in the Q band region. It suggests that the VOPc film is deposited with pseudorandomity. On the other hand, the VOPc film (Sample 2) on the substrate coated with 300°C has an absorption peak at 400nm in the Q band region.

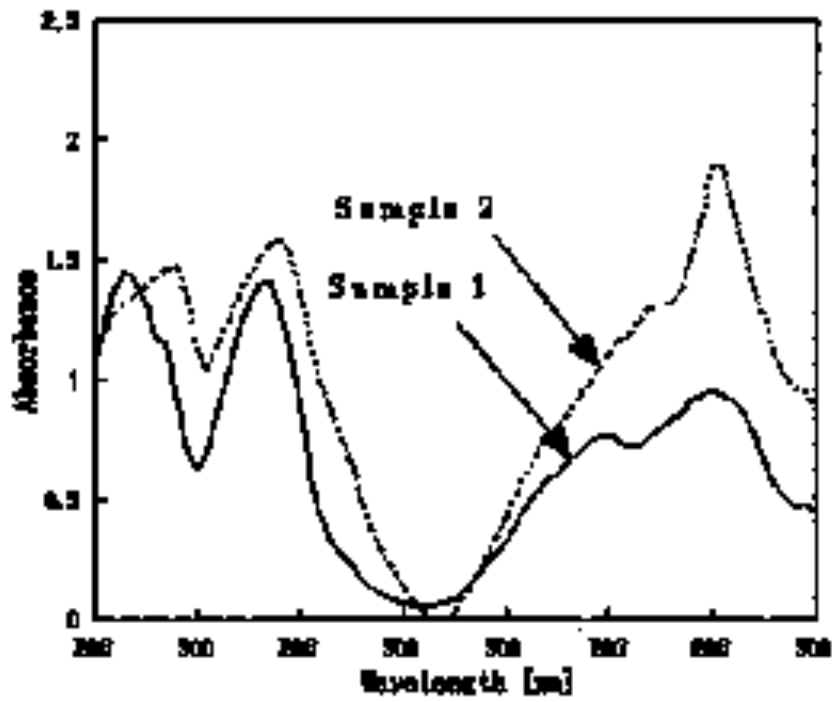


Fig. 1. VIS/UV spectra of Sample 1 and 2.



Fig. 2. SEM image of Sample 3.

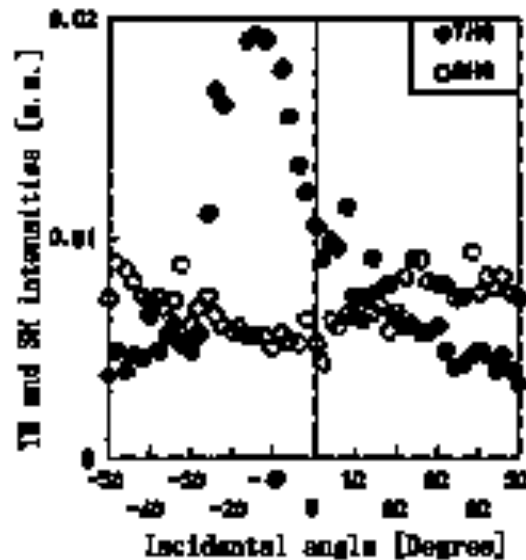


Fig. 8 SE and TE intensities vs. incidental angle of Sample 2.

It suggests that the VOPc film is closely related to the structure of epitaxy. The SEM image of Sample 1 shows a SE of mixed crystals. Its orientation has two axes. Figure 8 shows the SEM image of Sample 2. The mixed crystals are larger than those of Sample 1. From the distance between crystals of the MERRID pattern of Sample 2, VOPc structure is the film film grown on NaCl substrate from two locusts ($\sqrt{10}a_{\text{NaCl}}/20 \pm 37^\circ$). The second and third harmonic generations (TE and SE) of VOPc film were measured by X-ray diffraction. The incident angle dependence of SE intensity of Sample 1 irradiated with Φ polarized laser beam shows a V curve whose minimum value is located at 0 degree. It suggests that the structure in the VOPc film has a Φ angle on NaCl substrate. On the other hand, the incident angle dependence of TE intensity of Sample 1 irradiated with Φ polarized laser beam shows an upper convex curve that has a maximum value at -4 degree. Figure 8 shows the incident angle dependence of SE and TE intensity of Sample 2 irradiated with Φ and Ψ polarized laser beam. The SE intensity shows a symmetrical V curve at 0 degree but the SE intensity is not zero at 0 degree. It means that the VOPc film contains the structure parallel to substrate in VOPc film. The TE intensity of Sample 2 has an upper convex curve whose minimum value is located -16 degree. They suggest that the TE susceptibility tensor of VOPc film is dominated with the components of χ_{1111} and χ_{3333} and also indicate that the VOPc molecules deposited on NaCl have a Φ angle. These may give an information about TEH.

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