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分子線エピタキシー法によりNaCl基板上に作製されたバナジルフタロシアニン膜の3次高調波

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 suggest that the tensor of TH susceptibility of VOPc film is dominated with the components of χ(xxxx) and χ(yyyy) and also indicates that the VOPc molecules deposited on NaCl have a tilt angle. These may give us information about THG.

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 χ(3) is expected if appropriately oriented molecules can be manufactured in film form. It has been demonstrated that organic materials with π-conjugated systems show anomalously large optical nonlinearities and ultrafast 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 γ(ijkl)(-ω_4; ω_1, ω_2, ω_3) main chain axis component γ(xxxx))(-ω_4; ω_1, ω_2, ω_3) with all electric fields aligned along the chain axis (x-axis).
In changing from linear to cyclic conjugated structures, one must take new tensor components such as γ(yyyy), γ(xxyy), γ(xyxy) γ(xyyx), γ(yyxx), γ(yxyx) and γ(yxxy) into consideration. Therefore, the nonlinear optical properties of the organic thin films on thermally treated substrate were investigated.

2. Experiment

The source material used was VOPc powder supplied from Eastman Kodak Company. After VOPc powder was inserted into a Knudsen-cell, it was preheated at 300°C for one hour. The substrate of NaCl was cleaved just before a setting to a holder. After that, the substrate was attached on the holder. The main chamber of MBE was at about 10^{-7} Pa, and the NaCl substrate was preheated for one hour. The preheating was done at 150°C and 250°C. The evaporating temp. was kept at Te (evaporating temp.) : 300°C. The preparing conditions of VOPc thin films were the substrate temp. (Ts : 200°C), evaporating time (t: 240 min.), annealing temp. (Ta : 200°C) and annealing time (ta : 240 min.), respectively. The film thicknesses were 115nm for Sample 1 (preheating temp. of substrate: 150°C) and 130nm for Sample 2 (250°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 800nm in the Q band region. It suggests that the VOPc film is deposited with pseudo

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epitaxy. On the other hand, the VOPc film (Sample 2) on the substrate treated with 250°C has an absorption peak at 805nm in the Q band region.

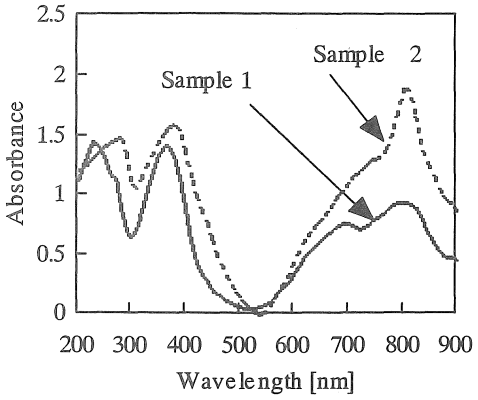

Fig.1 VIS/UV spectra of Sample 1 and 2.

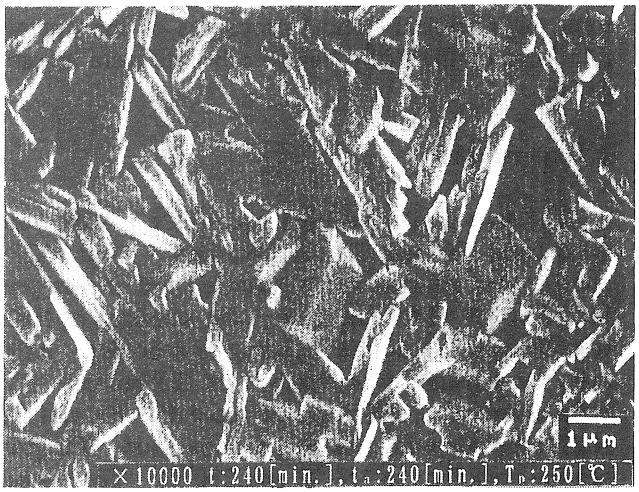

Fig. 2 SEM image of Sample 2.

It suggests that the VOPc film is closely related to the structure of epitaxy. The SEM image of Sample 1 shows a lot of island crystals. Its orientation has two axes. Figure 2 shows the SEM image of Sample 2. The island crystals are larger than those of Sample 1. From the distance between streaks of the RHEED pattern of Sample 2, VOPc molecules in the thin film grown on NaCl substrate form two lattices $(10)^{1/2}\times(10)^{1/2}\pm27^{o}$. The second and third harmonic generations (TH and SH) of VOPc films were measured by Maker fringe. The incident angle dependence of SH intensity of Sample 1 irradiated with S polarized laser beam shows a V curve whose minimum value is located at 5 degree. It suggests that the molecules in the VOPc film have a tilt angle on NaCl substrate.

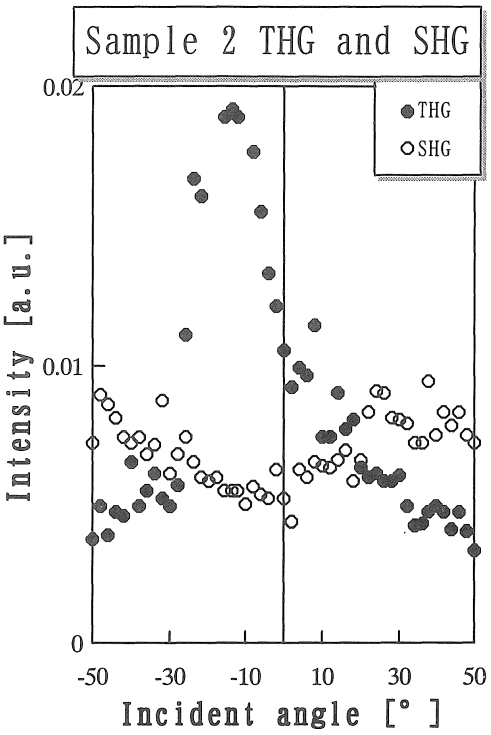

Fig.3 SH and TH intensities vs. incidental angle of Sample 2.

On the other hand, the incidental angle dependence of TH intensity of Sample 1 irradiated with P polarized laser beam shows an upper convex curve that has a maximum value at -5 degree. Figure 3 shows the incident angle dependence of SH and TH intensity of Sample 2 irradiated with S and P polarized laser beam. The SH intensity shows a symmetrical V curve at 0 degree but the SH intensity is not zero at 0 degree. It means that the VOPc film contain the molecules normal to substrate in VOPc film. The TH intensity of Sample 2 has an upper convex curve whose maximum value at is located -15 degree. They suggest that the TH susceptibility tensors of VOPc film is dominated with the components of χ(xxxx) and χ(yyyy) and also indicate that the VOPc molecules deposited on NaCl have a tilt angle. These may give us information about THG.

4. Conclusions

The VIS/UV spectrum of Sample 1 has an absorption peak at 800nm in the Q band region. It suggests that the VOPc film is deposited with pseudoepitaxy. On the other hand, the VOPc film (Sample 2) on the

substrate treated with 250^{O}C has an absorption peak at 805nm in the Q band region. It suggests that the VOPc film is closely related to the structure of epitaxy. We indicated that the VOPc molecules in the thin film grown on NaCl substrate formed two lattices$(10)^{1/2}\times(10)^{1/2}\pm27^{O}$ from the distance between streaks of the RHEED pattern of Sample 2. This paper was published by International conference of Nonlinear Optics Materials, Fundamentals and Applications held on 2000.

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