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Analysis of Second Harmonic Generation Phase Measurement for Determination of Molecular Orientation and Effective Second Order Nonlinear Optical Constant

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The average direction of molecules and the absolute value of an effective second order nonlinear optical constant ($|d^{eff}|$) were simultaneously determined using a new second harmonic generation (SHG) phase measurement with theoretical curve fitting. The theoretical curve fitting process was demonstrated using poled poly (methyl methacrylate) films doped with p-nitroaniline (PNA/PMMA), which were used as a reference and a sample. The second harmonic (SH) interference generated between the two poled PNA/PMMA films was modulated by rotating the phase generator inserted between the two. The fitting process derived d^{eff} itself. The average direction of molecules was shown by the sign of d^{eff}. The obtained value of $|d^{eff}|$ was verified by that from a Maker fringe measurement. The obtained average direction of molecules coincided with that determined by a poling geometry.

Keywords: thin film; phase measurement; interference; SHG; second order nonlinear optical constant

INTRODUCTION

Efficient second order nonlinear optical (NLO) property have been required for NLO application utilizing thin films. [1] To achieve that requirement, many researchers have focused on how to increase the

degree of polar alignment, and how to keep its stability as well as developing molecules possessing large first order hyperpolarizability. [2]-[4] Second harmonic generation (SHG) phase measurements are powerful tool to investigate molecular orientation. [2]-[5] In conventional SHG phase measurements, the average direction of molecules in a sample has been determined in comparison with that of a standard sample like a quartz crystal. In this paper, we will introduce a new method where the average direction of molecules is obtained by measuring only the sample without the standard sample. This method also provides the absolute value of an effective second order nonlinear optical constant ($|d^{eff}|$) for the sample. Both $|d^{eff}|$ and the average direction of molecules, or the sign of d^{eff}, are obtained by the theoretical curve fitting performed to the SH interfered curve.

EXPERIMENTAL

Poled poly (methyl methacrylate) films doped with 2 wt.% of p-nitroaniline (PNA/PMMA) were used as a reference and a sample. The direction of the dipole moment of PNA in PMMA was controlled by the corona poling. The poling geometry was shown in Figure 1.

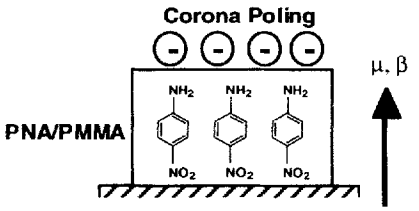

FIGURE 1 Poling geometry of PNA/PMMA film.

Figure 2 shows the optical setup of the new SHG phase measurement. The fundamental beam was a p-polarized Q-switched Nd: YAG laser, and then p-polarized SH signal was detected. The

fundamental beam was radiated on the reference film at an incident angle of 45°, and a certain amount of fundamental beam was converted to a SH beam by passing through the medium of the reference. The rest of the fundamental beam passed through the reference was radiated on the sample film at an incident angle of 45°. The reference and the sample SH beams interfere with each other. The interfered light power was modulated by rotating the phase generator inserted between the two films. The SH intensity was measured as a function of the rotating angle of the phase generator. The fitting program was applied to the modulated SH interference power curve. The accuracy of the fitting process was demonstrated by comparing the value of $|d^{eff}|$ for the sample with that determined by a Maker fringe measurement and also by comparing the average direction of molecules with that determined by the poling geometry. [6]

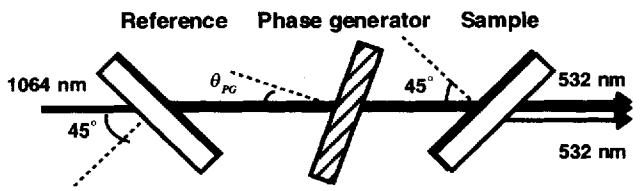

FIGURE 2 Optical setup of SHG phase measurement.

RESULT AND DISCUSSION

Figure 3 shows the SH interference fringe and the fitted curved. The fitted curve is in good agreement with the experimental data. The value of $|d^{eff}|$ for the sample was calculated by performing the curve fitting, and the average direction of molecules was determined by the sign of d^{eff}. $|d^{eff}|$ was obtained to be 0.3 pm/V. On the other hand, $|d^{eff}|$ of the sample at an incident angle of 45° was calculated to be 0.3 pm/V using values (d_{31}, d_{33}) obtained by Maker fringe measurement. The average

direction of molecules was obtained as a positive sign for the d^{eff}, showing that the sample had the same average direction of molecules with the reference. This shows the average direction of molecules in the sample is coincided with that determined by the poling geometry.

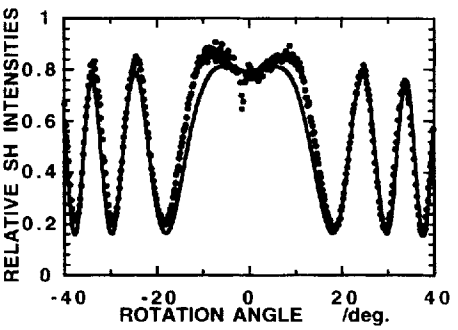

FIGURE 3 SH interference fringe (dots) and the fitted curve (solid line).

CONCLUSION

We have shown analysis of SHG phase measurement for determination of molecular orientation and the value of $|d^{eff}|$. The fitting curve was in good agreement with the experimental data.

The new SHG phase measurement and the curve fitting program can be widely used to effective method for investigation of NLO molecules.

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