Theoretical Investigation of Carrier Envelope Phase Dependence of All-Optical Poling with the Third- and Higher-Order Nonlinear Processes†

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Dependence of efficiency in all-optical poling with nonlinear processes, up to eighth order, is considered. The explicit form of the nonlinear susceptibility that is responsible for the poling is derived, which shows both CEP and phase mismatch dependence. On the basis of an analysis of pulse propagation in a nonlinear material, it is shown that one can identify the order of nonlinearity that is relevant to the poling process, relying on current technology of CEP stabilization and thin-film growth.

Introduction

All-optical poling is known to be an efficient orientation technique that is thought to be a promising candidate for a novel device of frequency conversion. It is realized in glass optical fibers,[1,2] dye solutions,[3] and spin-coated films.[4] The key element of the technology is the possibility of controlling polar molecular order with full optical means, leading to a spatially periodic structure capable of frequency doubling.

A new feature in laser technology, control of the carrier envelope phase (CEP), has attracted much attention and has been applied to the quantum interference experiment.[5−8] A novel way of active control of the CEP was achieved based on the interference-feedback method.[9] Another type of CEP control, the passive control method, is realized based on optical parametric amplification.[10] Furthermore, a hybrid type of CEP control was proposed and demonstrated.[11] Among them, a self-stabilized CEP was achieved using an idler from an optical parametric amplifier (OPA) pumped with the second harmonics of the Ti:Sapphire radiation and seeded with a white-light continuum.[12] Application of the CEP-controlled pulse has also been extensively studied, including the development of a soft-X-ray pulse source that is very important in medicine and industry.[13] The CEP-controlled light source was also used to demonstrate CEP-dependent efficiency in optical poling.[14]

Theoretical and experimental investigations of all-optical poling have been mainly based on a poling mechanism in which the writing process of the nonlinear susceptibility in a material is realized with the third-order nonlinear process. The nonlinear susceptibility is formed by the quantum interference between two-photon absorption of the ω-frequency field and one-photon absorption of the 2ω-frequency component. In the optical poling experiment with a controlled CEP,[14] however, it is known that the sample used has a number of excited states that can contribute to the poling process. One can hypothesize that the writing is done with a higher-order, rather than the third-order, nonlinear process. It is even needed to take into account the optical poling with such higher-order nonlinear process if the sample under consideration is transparent for two-photon absorption of the ω-frequency field and one-photon absorption of the 2ω-frequency one. Experimental observations, indeed, have been made for optical poling involving fifth-order nonlinearity, in which the origin of the photoinduced $\chi^{(2)}$ was attributed to simultaneous three-photon ($\omega + \omega + \omega$) and two-photon ($2\omega + \omega$) processes.[15,16] The aim of this paper is to consider the CEP dependence of the higher-order nonlinear poling. A methodology to clarify which order of nonlinearity is relevant to the poling process under consideration is investigated.

CEP Dependence of the Nonlinear Susceptibility

In all-optical poling, simultaneous irradiation of the fundamental wave (E_ω) and its second harmonic (SH) wave ($E_{2\omega}$) leads to the formation of a polar field that is described by the coherent superposition of these fields and characterized by its nonzero temporal average $\langle E^3\rangle_t \neq 0$; $E = E_\omega + E_{2\omega}$ depending on the relative phase between the two. A quasi-permanent poling induced by this field breaks the centrosymmetry in the sample. The nonlinear susceptibility $\chi^{(2)}_{\mathrm{ind}}$ caused by this coherent nonlinear interaction is known to be proportional to $\langle E^3\rangle_t$. Therefore, the SH generation through this $\chi^{(2)}_{\mathrm{ind}}$ is considered to be a six-wave mixing (SWM) process and has been extensively studied.[17−23]

The output of the noncollinear optical parametric amplifier (NOPA), which is used as a CEP-controlled light source in an optical poling experiment,[14] is characterized by its ultrabroadband spectral feature that covers nearly the whole visible range. The self-stabilized carrier envelope phase (CEP) was also demonstrated experimentally. Orientational hole burning was performed by a selective excitation of molecules whose transition dipoles were parallel or antiparallel to the polarization of the writing field, using a sample film of a tetrahydrofuran solution of 4-(*N*-(2-hydroxyethyl)-*N*-ethyl)-amino-4′-nitroazobenzene [disperse red 1 (DR1)] and poly(methyl methacrylate) (PMMA) spin coated on a glass substrate.[14] A macroscopic

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dipole was generated due to subsequent trans−cis−trans isomerization that led to a decrease of molecules with specific orientation. The CEP dependence of $\chi^{(2)}_{ind}$ is generally given as follows

$$\chi^{(2)}_{ind} \propto \chi^{(2)}_{eff} \cos[\Delta k + \Delta\phi + (\phi_2^{CE} - 2\phi_1^{CE})] \qquad (1)$$

with $\chi^{(2)}_{eff} \equiv \alpha\left|\left|(\tilde{E}^0_\omega)^2 \tilde{E}^{0*}_{2\omega}\right|\right|$.

The $\Delta k \equiv k_2 - 2k_1$ is the wave-vector mismatch, where k_i's ($i = 1, 2$) are the wavenumber of the fundamental field and SH, respectively, $\Delta\phi$ is the relative phase between these fields, and ϕ_i^{CE} ($i = 1, 2$) are the carrier envelope phases of the fundamental field and SH, respectively. $\tilde{E}^0_\Gamma$ ($\Gamma = \omega, 2\omega$) are the complex amplitudes of the fundamental field and SH, respectively, and α is a proportionality constant. In this experiment, $\chi^{(2)}_{ind}$ is induced by a nonlinear process through interference between the ω and 2ω components of the NOPA output, which is a broadband frequency comb spanning nearly one octave. The readout process is based on the SH generation of the fundamental which is used also in the writing stage. In the poling experiment using a CEP-controlled light source,[14] however, it is expected that an even higher order nonlinear process is responsible for the formation of $\chi^{(2)}_{ind}$, concerning the laser spectrum and the resonance energy of the S_1 of the molecule. Motivated with this consideration, the CEP-dependent behavior of $\chi^{(2)}_{ind}$ induced by the fourth- and higher-order nonlinear processes, together with relevant SH, is discussed. The beam configuration under consideration is one-dimensional. Hereafter, only the propagation coordinate is taken into account as the position degree of freedom, ignoring the radial distribution of electric fields and hence induced nonlinear susceptibility.

First, let us consider $\chi^{(2)}_{ind}$ induced by the fifth-order process. Here, $\chi^{(2)}_{ind}$ is proportional to $\langle E^5\rangle_t$. In this case, the whole write and read process is an eight-wave mixing process. With our notation

$$E_\omega(z, t) = \tilde{E}_\omega + \tilde{E}^*_\omega = \tilde{E}^0_\omega \exp(-i\omega t - ik_1 z + i\phi_1^{CE}) + \text{c.c.}$$

and

$$E_{2\omega}(z, t) = \tilde{E}_{2\omega} + \tilde{E}^*_{2\omega} = \tilde{E}^0_{2\omega} \exp(-i2\omega t - ik_2 z + i\phi_2^{CE} + i\Delta\phi) + \text{c.c.}$$

it leads

$$\langle E^5\rangle_t = \langle (E_\omega(z, t) + E_{2\omega}(z, t))^5\rangle_t \propto 2(\tilde{E}^3_\omega \tilde{E}^*_\omega \tilde{E}^*_{2\omega} + \text{c.c.}) + 3(\tilde{E}^2_\omega \tilde{E}_{2\omega} \tilde{E}^{2*}_{2\omega} + \text{c.c.}) \qquad (2)$$

The symmetry breaking $\langle E^5\rangle_t$ is composed of two terms. The first one is proportional to the quadratic power of the frequency ω component times the linear field of the frequency 2ω component. The second one is proportional to a product of the powers of the ω and 2ω components times the linear field of the 2ω component. Both terms are relevant to interferences between two- and three-photon processes, and resonant frequencies of the excited states relevant to the first and the last terms are 3ω and 4ω, respectively. If we define $\chi^{(2)}_{eff}$ such as

$$\chi^{(2)}_{eff} \equiv 2(\tilde{E}^0_\omega)^4 \tilde{E}^0_{2\omega} + 3(\tilde{E}^0_\omega)^2 (\tilde{E}^0_{2\omega})^3$$

The SH intensity I^{SH} is given as

$$I^{SH} \propto I^2_\omega (\chi^{(2)}_{eff})^2 \cos^2(-\Delta kz + \phi^{CE} - \Delta\phi) \qquad (3)$$

TABLE 1: CEP Dependence of I^{SH} for Several Orders

orders of the $\chi^{(2)}$ induction nonlinear process	$\Delta\phi^{CE}$ dependence of I^{SHG}
4	$\propto I^2_\omega (\chi^{(2)}_{eff})^2$ (no dependence on ϕ^{CE})
5	$\propto I^2_\omega (\chi^{(2)}_{eff})^2 \cos^2(-\Delta kz + \phi^{CE} - \Delta\phi)$
6	$\propto I^2_\omega \{\chi^{(2)}_{eff}[\cos 2(-\Delta kz + \phi^{CE} - \Delta\phi) + 12] + \chi^{(2)}_{const}\}$
7	$\propto I^2_\omega (\chi^{(2)}_{eff})^2 \cos^2(-\Delta kz + \phi^{CE} - \Delta\phi)$
8	$\propto I^2_\omega (\chi^{(2)}_{eff})^2 \cos^2 2(\Delta kz - \phi^{CE})$

where I_ω is the reading field intensity. The CEP $\phi^{CE} \equiv \phi_1^{CE} = 2\phi_1^{CE} - \phi_2^{CE}$ (because $\phi_1^{CE} = \phi_2^{CE}$ for NOPA output) is employed. The CEP dependence is the same as that in the case of SWM. It is shown that the CEP dependence of $\chi^{(2)}_{ind}$ and I^{SH} (the SH intensity) are identical to the one in SWM. The CEP dependence of I^{SH} for several orders are summarized in Table 1. Here, $\chi^{(2)}_{const}$ ($=12(\tilde{E}^0_\omega)^2(\tilde{E}^0_{2\omega})^4$) is a constant independent of CEP. I^{SH} in units of $[I_\omega(\chi^{(2)}_{eff})]^2$ is plotted as a function of CEP with the proportionality constant set to one (Figure 1). The phase matching condition $\Delta k = 0$ was employed, which is valid for our system in which the coherent length of the sample is much larger than its thickness.[14] The relative phase $\Delta\phi$ was also assumed to be zero. A relation $\chi^{(2)}_{const} = 12\chi^{(2)}_{eff}$ was employed, which holds provided that $\tilde{E}^0_\omega$ and $\tilde{E}^0_{2\omega}$ are identical, which is approximately correct for the NOPA output. It is shown that the order of nonlinearity relevant to the SHG can be identified by monitoring the intensity variation of the SH as a function of the CEP. Photoinduction of $\chi^{(2)}_{ind}$ with the third-, fifth-, or seventh-order nonlinear processes results in the identical CEP dependence in $\chi^{(2)}_{ind}$ and, hence, in I^{SH}. It is impossible to attribute the poling process to one of these orders that is responsible for the SH under consideration only from the CEP dependence of I^{SH}. Photoinduction through a fourth-order process, on the other hand, is independent of the CEP. It is found that, even when $\langle E^3\rangle_t = 0$ holds and $\Delta k = 0$ is properly assumed (i.e., the position dependence in $\chi^{(2)}_{ind}$ is negligibly small), $\chi^{(2)}_{ind}$ may not be induced, depending on CEP. As a fact, $\chi^{(2)}_{ind}$ and hence I^{SH} are zero at $\phi^{CEP} = (\pi/2)n$, ($n = 1,2, ...$). If, on the other hand, the fourth-order process contributes to the induction of $\chi^{(2)}_{ind}$, one would not have such ϕ^{CEP}'s at which $\chi^{(2)}_{ind}$ disappears. In the case of the sixth-order process, I^{SH} shows a CEP dependence similar to that in the case of the third-order, but the dynamic range is substantially smaller than the absolute value of I^{SH}. In the case of the eighth-order, the frequency of the oscillation with respect to the CEP is twice as large as that

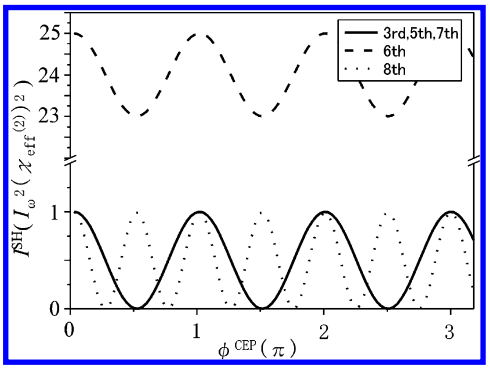

Figure 1. I^{SH} in units of $I^2_\omega(\chi^{(2)}_{eff})^2$ as a function of the CEP with the proportionality constant set to one; $\Delta k = 0$ is assumed, which is consistent with the experiment under consideration, in which the coherent length of the sample is much larger than its thickness.

in the case of the third-order. Thus, one can tell which order of nonlinearity contributes to the dynamically induced SH by investigating the CEP-dependent behavior of its intensity, even though there is a limitation, that is, the fifth- and seventh-order processes are not distinguishable from the third-order one. Note that from the $\chi^{(2)}_{ind}$ dependence on the product of the ω- and 2ω-frequency field amplitudes, which was measured in terms of either the initial growth of $\chi^{(2)}_{ind}$ or saturated SHG,[15,16] one can consider the order of the nonlinear process that is relevant to the $\chi^{(2)}_{ind}$ induction. Since the measurement error is inevitable, the CEP and the product of the amplitude dependence of $\chi^{(2)}_{ind}$ can be used complementarily to determine the nonlinear order more definitely.

Sample Thickness Dependence of Poling Efficiency

Next, let us consider the dynamic process of SH generation through an interaction of the reading pulse propagating through media with nonlinear polarization generated by the optical poling. The SH generation process is explicitly treated based on a differential equation expressing the field generation due to the polarization, and the SH intensity as a function of the CEP as well as the sample thickness is considered to investigate the possibility of identifying the order of the nonlinearity inducing poling. The differential equation to be solved is

$$\frac{\partial^2}{\partial z^2}E_{SH}(z,t) - \frac{n_2^2}{c^2}\frac{\partial^2}{\partial t^2}E_{SH}(z,t) = \mu\frac{\partial^2}{\partial t^2}P^{NL} \quad (4)$$

Here, E_{SH} is the electric field of the generated SH, P^{NL} is the nonlinear polarization written in the sample through the optical poling process, μ and c are the permeability and light speed in the vacuum, respectively, and n_2 is the refractive index of the sample under consideration for the SH field. Similar to the poling experiment, polarization of both the SH field and the writing field, together with P^{NL}, is assumed to be in the same direction. In this case, field variables and polarization can be treated as scalars. In the poling experiment, the width of the reading pulse is as short as 40 fs. In order to include a dispersion relation, the pulse electric field is written in the form of integrated plane-wave components, that is

$$E(z,t) = \int E'(\omega)\exp[-i(\omega t - k(\omega)z)]\mathrm{d}\omega \quad (5)$$

Here, $E(z,t)$ satisfies the Maxwell's equation as far as $E'(\omega)$ is a solution of the equation since the Maxwell's equation is linear. In such a treatment, however, the equation under consideration turns out to be fairly complex. In our study, consequently, the writing and the SH fields are assumed to be written in the following forms

$$E_\omega = \tilde{E}^0_\omega \exp[-i(\omega_1 t - k_1 z)]\exp\left[-\frac{(z - v_1^g t)^2}{\Delta^2}\right] \quad (6a)$$

and

$$E_{SH} = \tilde{E}^0_{SH} \exp[-i(\omega_2 t - k_2 z)]\exp\left[-\frac{(z - v_2^g t)^2}{\Delta^2}\right] \quad (6b)$$

In this formulation, the dispersion effect is taken in account in a limited way, that is, refractive indices, n_i's, and hence k_i's ($i = 1, 2$) for both fields are constant in the frequency region that is covered by each pulse. The v_i^g ($i = 1, 2$) are the group velocity of the writing field and SH, and Δ is the pulse width in z-space. Furthermore, additional third-order nonlinear effects, such as self-phase modulation, are assumed to be negligibly small.

The nonlinear polarization is, therefore

$$P^{NL} = \epsilon\chi^{(2)}_{ind}(\tilde{E}^0_\omega)^2 \exp[-i2(\omega_1 t - k_1 z)]\exp\left[-\frac{(z - v_1^g t)^2}{\Delta^2}\right] \quad (7)$$

where ϵ is the dielectric permittivity. It follows

$$\mu\frac{\partial^2}{\partial t^2}P^{NL} = \mu\epsilon\chi^{(2)}_{ind}(\tilde{E}^0_\omega)^2\left\{\left[\frac{4v_1^g}{\Delta^2}(z - v_1^g t) - i2\omega_1\right]^2 - \frac{4(v_1^g)^2}{\Delta^2}\right\} \times \exp[-i2(\omega_1 t - k_1 z)]\exp\left[-\frac{2(z - v_1^g t)^2}{\Delta^2}\right] \quad (8)$$

The optical density for the wavelength region under consideration is shown to be very small in the poling experiment, and the time derivative of the electric field of the incident pulse, consequently, is neglected.

Employing the slowly varying envelope approximation, the second derivatives of E_{SH} are

$$\frac{\partial^2}{\partial z^2}E_{SH}(z,t) = \left\{2\left[ik_2 - \frac{2}{\Delta^2}(z - v_2^g t)\right]\left(\frac{\partial}{\partial z}\tilde{E}^0_{SH}\right) + \left[ik_2 - \frac{2}{\Delta^2}(z - v_2^g t)\right]^2 \tilde{E}^0_{SH} - \frac{2}{\Delta^2}\tilde{E}^0_{SH}\right\} \times \exp[-i(\omega_2 t - k_2 z)]\exp\left[-\frac{(z - v_2^g t)^2}{\Delta^2}\right] + \text{c.c.} \quad (9a)$$

and

$$\frac{\partial^2}{\partial t^2}E_{SH}(z,t) = \left\{-2\left[i\omega_2 - \frac{2v_2^g}{\Delta^2}(z - v_2^g t)\right]\left(\frac{\partial}{\partial t}E^0_{SH}\right) + \left[i\omega_2 - \frac{2v_2^g}{\Delta^2}(z - v_2^g t)\right]^2 \tilde{E}^0_{SH} - \frac{2(v_2^g)^2}{\Delta^2}\tilde{E}^0_{SH}\right\} \times \exp[-i(\omega_2 t - k_2 z)]\exp\left[-\frac{(z - v_2^g t)^2}{\Delta^2}\right] + \text{c.c.} \quad (9b)$$

In our system, $(v_2^g/\Delta^2)(z - v_2^g t)$ and $(z - v_2^g t)/\Delta^2$ are found to be 3 orders of magnitude smaller than ω_2 and k_2, respectively. It follows that

$$\frac{\partial^2}{\partial z^2}E_{SH} = \left[i2k_2\left(\frac{\partial}{\partial z}\tilde{E}^0_{SH}\right) - (k_2)^2\tilde{E}^0_{SH} - \frac{2}{\Delta^2}\tilde{E}^0_{SH}\right] \times \exp[-i(\omega_2 t - k_2 z)]\exp\left[-\frac{(z - v_2^g t)^2}{\Delta^2}\right] + \text{c.c.} \quad (10a)$$

and

$$\frac{\partial^2}{\partial t^2}E_{SH} = \left[-i2\omega_2\left(\frac{\partial}{\partial t}\tilde{E}^0_{SH}\right) - (\omega_2)^2\tilde{E}^0_{SH} - \frac{2(v_2^g)^2}{\Delta^2}\tilde{E}^0_{SH}\right] \times \exp[-i(\omega_2 t - k_2 z)]\exp\left[-\frac{(z - v_2^g t)^2}{\Delta^2}\right] + \text{c.c.} \quad (10b)$$

Assuming that the amplitude of the field depends only on z, the equation is obtained as

$$\frac{\partial}{\partial z}\tilde{E}^0_{SH} - \frac{i}{\Delta^2 k_2}\left[\left(\frac{v_2^g n_2}{c}\right)^2 - 1\right]\tilde{E}^0_{SH} = -i\frac{\mu\epsilon}{2k_2}\chi^{(2)}_{ind}(\tilde{E}^0_\omega)^2\left\{\left[\frac{4v_1^g}{\Delta^2}(z - v_1^g t) - i2\omega_1\right]^2 - \frac{4(v_1^g)^2}{\Delta^2}\right\}\exp(-i\Delta k z)\exp\left\{-\frac{1}{\Delta^2}[2zt(v_2^g - 2v_1^g) - ((v_2^g)^2 - 2(v_1^g)^2)t^2 + z^2]\right\} \quad (11)$$

Here, $\Delta k \equiv k_2 - 2k_1$ is the phase mismatch, and $k_2 = (n_2\omega_2)/c$ is employed. This is solved as

$$\tilde{E}^0_{SH}(z) = \exp\left\{\frac{i}{\Delta^2 k_2}\left[\left(\frac{n_2 v_2^g}{c}\right)^2 - 1\right]z\right\} \times \int_0^z -i\frac{\mu\epsilon}{2k_2}\chi^{(2)}_{ind}(\tilde{E}^0_\omega)^2\left\{\left[\frac{4v_1^g}{\Delta^2}(z' - v_1^g t) - i2\omega_1\right]^2 - \frac{4(v_1^g)^2}{\Delta^2}\right\}\exp(-i\Delta k z')\exp\left\{-\frac{1}{\Delta^2}[2z't(v_2^g - 2v_1^g) - ((v_2^g)^2 - 2(v_1^g)^2)t^2 + z'^2]\right\} \exp\left\{-\frac{i}{\Delta^2 k_2}\left[\left(\frac{n_2 v_2^g}{c}\right)^2 - 1\right]z'\right\}dz' \quad (12)$$

The sample thickness, z, used in the experiment was 1 μm, which is $\sim 0.6(2\pi/k_1)$ for the reading field of which the wavelength is 1600 nm. In this study, however, z values up to 20 μm are considered to investigate the z dependence of the SH intensity. The pulse width is 40 fs $= 8(2\pi/\omega_1)$. The ratio of the refractive indices, n_2/n_1, is 1.13, which is also in accordance with the experiment. Group velocities in the unit of phase velocity of the reading field, v_1^p, are $v_1^g = 1.5v_1^p$ and $v_2^g = 0.65\ v_1^p$, which are estimated from the dispersion curve obtained in the experiment. It is found that although both temporal and spatial behaviors of the SH intensity show little dependence on the CEP change, they vary substantially due to the change in n_2/n_1 and the relation between group velocities of the fields. The SH intensity that is effected by n_2/n_1 and the group velocities will be discussed elsewhere. In the present study, the discussion is limited to the dependence on the sample thickness in which the refractive indices and group velocities are fixed to maintain the experimental condition. The SH intensity integrated over the temporal range of $-10-15(2\pi/\omega_1)$ at each thickness of the sample is shown in Figure 2. Here, the origin of the time axis is the moment at which the peak of the incident reading pulse reaches to the surface of the sample. The SH intensity as a function of sample thickness is effected by the periodic oscillation due to the phase mismatch, Δk, the z dependence of $\chi^{(2)}_{ind}$, and the alternative SH and fundamental wave generation in which those fields are generated alternatively due to the second-order nonlinear process. It is seen that, from Figure 2, the z dependence of the SH intensity that is typical in each order of poling is seen in a region roughly larger than $z \approx 5(2\pi/k_1)$. Behind this point, let us term this point a critical point, the SH intensity is dominated by generation at the beginning stage ($0 \le z \le 2$) and a subsequent stage of decay. In the overlapping region, which is the region behind the critical point, the envelope of the incident pulse has a considerable overlap with the envelope of the SH, leading to the enhanced SH generation. Beyond the critical point, this overlap reduces due to the difference in group velocities between the incident fundamental wave and the SH. Let us estimate roughly to what extent the incident pulse "substantially" overlaps with that of the SH. The difference in the group velocities is $0.9v_1^p$. The time needed for two pulses, the incident and SH pulses that initially perfectly overlap each other to separate with a distance of half of the pulse width, $4(2\pi/k_1)$, is

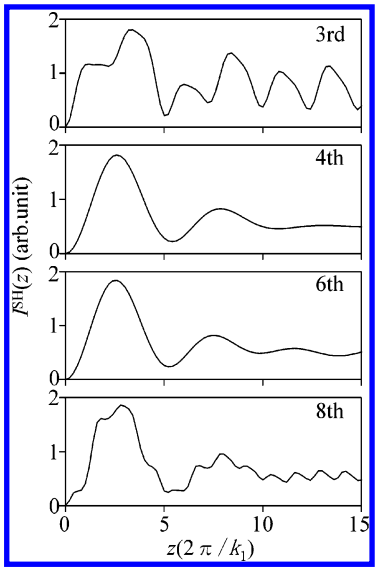

Figure 2. Temporally integrated SH intensity at each thickness of the sample.

$$\frac{4\dfrac{2\pi}{k_1}}{0.9\dfrac{\omega_1}{k_1}} = 4.4\frac{2\pi}{\omega_1} \quad (13)$$

Therefore, roughly speaking, the overlap is large at most $z = 4.4(2\pi/k_1)$. This corresponds to the position of the critical point. It is concluded that one needs to investigate the sample thickness dependence of the SH intensity beyond the critical point that is determined by the difference in group velocity to identify which order is relevant to the optical poling.

Conclusion

In the poling experiment, the CEP dependence of the SH intensity affects the efficiency of the writing process, which is observed as the rate of growth of the SH intensity. It is possible, in principle, to determine the order of nonlinearity relevant to the poling process from the observation of the rate of growth with changing CEP. Current technology in CEP stabilization enables such a measurement. In the present consideration, with formulating of the dynamic process of SH generation inside of the nonlinear medium, the order determination is shown to be possible from the SH intensity dependence on the sample thickness. The effect of change in the refractive index, group velocity, and self-phase modulation is a matter of future study.

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