



Carrier-envelope phase measurement by all-optical poling with a polar side-chain polymer

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ABSTRACT

Dependence of all-optical poling efficiency on carrier-envelope phase (CEP) could be measured using photoisomerization of dye molecules which are covalently bound to a polymer main chain and have large difference in static dipole moment between the ground state and excited state. Increased chromophore density leads to an order of magnitude reduction in signal-detection time from a dye doped polymer. Analysis of all-optical poling experiments with CEP changes clearly showed the presence of polarization restoring force to zero polarization. This enables resetting of SH activity in the all-optical poling process to be used for fast response loop of CEP stabilization. Phenomenological model could explain well the difference in the growth-and-decay dynamics of poling between sample of dye doped in polymer studied previously and that grafted to a polymer main chain used in the present paper.

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Control of relative phase between the carrier wave and the pulse envelope (carrier-envelope phase, CEP) of an ultrashort laser pulse is of vital importance for several applications such as soft X-ray generation [1] or frequency metrology [2,3]. Development of the CEP-measurement method began for amplifier outputs with the use of extreme nonlinear optical processes such as above threshold ionization (ATI) [4], which are sensitive to the field strength rather than the pulse intensity. However, because of the requirement of high field intensity, it is difficult to apply these methods to much weaker oscillator outputs. A compact experiment setup is difficult to realize in an ATI experiment because it requires a bulky vacuum system. Methods applicable with lower power in an atmospheric environment are preferable to be used for more compact CEP stabilized systems.

CEP measurement methods using quantum interference in condensed matter have such desired characteristics and are worth research. Quantum interference of injected photocurrent in

semiconductors has already been experimentally demonstrated for CEP measurement [5] and already utilized for stabilizing CEP of a laser system [6]. Another CEP dependent quantum interference utilizing all-optical poling was proposed in our group and demonstrated using azo-aromatic dye doped in a polymer [7]. In this paper, we show the clear CEP dependence of all-optical poling efficiency, an order of magnitude reduction of signal-detection time, and the presence of polarization restoring force in polar side-chain polymer that may be utilized for all optical erasure of induced polarization in the CEP measurement loop for a novel CEP stabilization scheme based on the all-optical poling.

The mechanism of all-optical poling is explained as follows. Azo-dye molecules, which can be efficiently photoisomerized, are irradiated by optical pulses simultaneously with frequency of ω and 2ω [8,9]. The quantum interference between the three-photon ($\omega + \omega + \omega$) absorption process and the two-photon ($\omega + 2\omega$) process leads to the inversion-symmetry-breaking asymmetry in the molecular excitation probability. The CEP dependence of the yield is the same as that of with two-photon ($\omega + \omega$) and one-photon (2ω) processes [10].

Mechanisms of all-optical poling is summarized as follows using the best known optical poling molecule, disperse-red 1 (DR1). Initially, *trans* form DR1 photoexcites and isomerizes to

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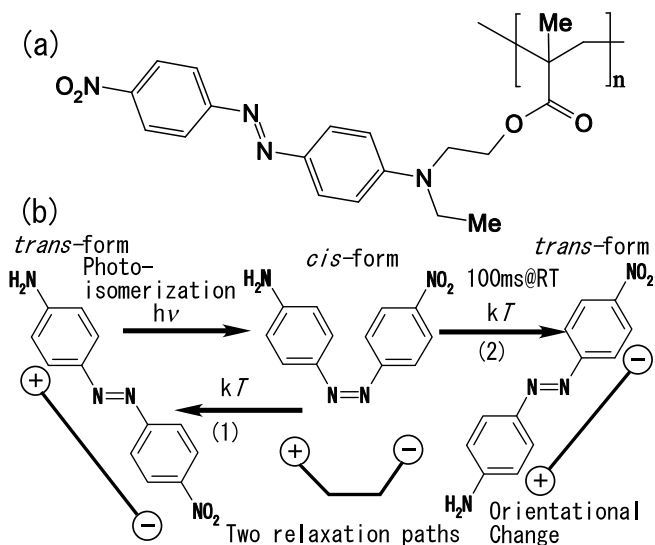


Fig. 1. (a) Poly(disperse red 1 methacrylate); (b) mechanism of all-optical poling utilizing *trans-cis-trans* photoisomerization cycle.

cis form, from which thermal (dark) isomerization back to *trans* form takes place. About 50% of the former molecules isomerize into the *trans* form with original orientation (channel (1)). The residual 50% can maintain the different orientation from the initial geometry (*Trans-cis-trans* photoisomerization cycle, Fig. 1b). Therefore, repeated asymmetric excitation causes asymmetric de-population in molecular orientation, which results in net poling in the whole molecular ensemble [11].

Because the order and the sign of molecular excitation asymmetry depend on the CEP of light pulses used for excitation [7], CEP determination can be performed by measuring the degree of molecular polarization, can be obtained from the efficiency of second harmonic generation (SHG).

The light source used for the experiment was the idler pulse from a non-collinear optical parametric amplifier (NOPA) [12–18]. The pump beam of the NOPA was the second harmonic (400 nm) of a Ti:sapphire regenerative amplifier pulse. A small fraction of the 400 nm beam was split off for white light generation in a CaF₂ plate and generated white light seeded the signal light to the NOPA. Because the pump and the signal beams of the NOPA had the same CEP, CEP of the idler is automatically stabilized [15,16,18]. In a real system, because there was small random drift in idler CEP, which was found to be caused mainly by air flow, the effect was corrected by negative feedback using the output of relative CEP measurement with *f*-2*f* spectral interferometry [19].

The spectrum of idler output is extending from 800 to 1600 nm with more than one octave. The second harmonic of idler is also present and output collinearly and simultaneously with the idler but in different polarization. We utilized two small spectral portions of idler pulse at 1600 nm and 800 nm as the ω light and the 2ω light sources, respectively, for the all-optical poling process.

The SHG efficiency measurement for reading the amount of poled molecules was performed by irradiating the sample only with 1600 nm light and detecting generated 800 nm light with a photomultiplier tube (PMT) and a lock-in amplifier. An 800 nm-bandpass glass filter was placed in front of the PMT to block 1600 nm light. Both beams were collinearly aligned and transmitted through a CaF₂ convex lens (focal length: 50 mm) to be focused on the surface of the sample.

Spin-coated chloroform solution of poly(disperse red 1 methacrylate) (Fig. 1a) on a glass substrate was used in poling experiment for CEP determination. In comparison with previous work [7],

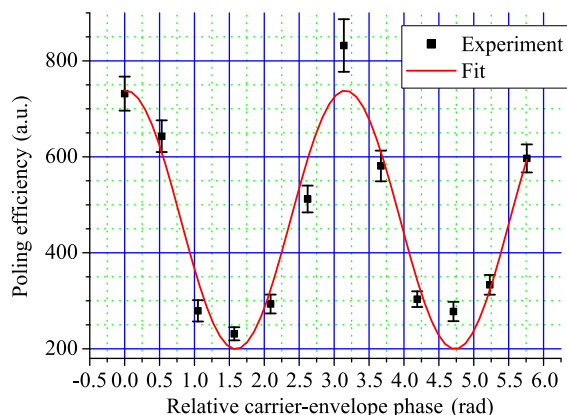


Fig. 2. All-optical poling efficiency as a function of relative carrier-envelope phase. Line shows a theoretical fit with $A + B \cos 2(\phi + \phi_0)$. $A = 470$, $B = 270$. The error bar of each point represents the standard deviation of parameter for each fitting of the SHG intensity in the each experimental run with linear increase with time.

which used DR1-doped poly-methylmethacrylate, 100%-dye-grafted polymer enabled us to achieve higher concentration of chromophores (82 wt%). The optical density of the sample probed at 500nm was 5.0, which was about two times higher than that of the previous work. The experiment was performed in the three independent experiments explained as follows.

In the first experiment, we studied the dependence of all-optical poling efficiency upon the CEP. All-optical poling experiment runs were performed with constant relative CEP. Each run consisted of 10–20 poling cycles. One poling cycle consisted of 15s poling and 2.5s SH intensity measurement. The SH intensity change in each run could be fit well with linear increase and the slope of the curve of SH intensity against runtime was defined as the all-optical poling efficiency η . Twelve runs of poling were performed at different sample positions with different relative CEPs and all-optical poling efficiencies for each run were plotted against relative CEP (Fig. 2). The whole data set could be well fit with a theoretical $\cos(2(\phi_{\text{rel}} - \phi_0))$ curve, in which ϕ_{rel} : relative CEP, ϕ_0 : initial CEP. The baseline offset from zero is due to the phase mismatch in finite thickness of the sample. Visibility C defined as $C = (\eta_{\text{max}} - \eta_{\text{min}}) / (\eta_{\text{max}} + \eta_{\text{min}})$ was 0.57 ± 0.05 . Sample thickness estimated from optical density of the sample is $1.6 \mu\text{m}$ and visibility estimated from the sample thickness was 0.5 ± 0.1 , which agrees with the experimental value. The time required to integrate SHG signal with signal to noise ratio higher than ten is about 5 min, which is an order of magnitude smaller than that of previous work [7], in which polymer film doped with DR1 was used as the poling sample.

In the second experiment, we studied SHG efficiency change upon the abrupt change of CEP. Hereafter, the term “SCR” is used to designate the stepwise change of relative CEP by π radian. We performed all-optical poling experiment runs almost the same as that of the first experiment. The difference existed in that the runs were longer (20–30 poling cycles) and the presence of SCR in the middle of each run. A typical SHG intensity change plotted as a function of exposure time is shown in Fig. 3. Before SCR, SHG intensity changes were the same as that of the first experiment and increased nearly linearly. Immediately after SCR, SHG intensity decreased to almost zero in less than 2 poling cycles. After several (4–5) poling cycles, SHG intensity resumed linear increase. The slope of increase after SCR is the same as that of before change within experimental error. To investigate the irradiation time dependence observed in the second experiment, we used a simple phenomenological model that describes the amount of poling. The model is a classical mass-and-spring model with friction force that

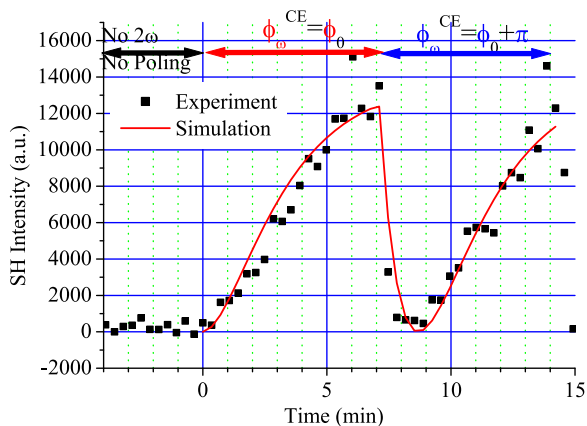


Fig. 3. SH intensity change during the all-optical poling experiment with the stepwise change of relative carrier-envelope phase by π radian.

is linear against translation speed. The phenomenological equation for the motion of the degree of polarization is given as,

$$dP(t)/dt = \alpha \cos(\phi) - \beta P(t). \quad (1)$$

Here $\alpha \cos(\phi)$ is CEP(ϕ)-dependent poling force, and $\beta P(t)$ is polarization-restoring force of the spring to zero poling, which is proportional to the current degree of polarization. The equation was solved numerically to obtain P^2 , which is proportional to the SH intensity. The model reproduced experimental curve well including the sharp decrease immediately after SCRP (Fig. 3).

In the third experiment, to check the validity of the model, an all-optical poling experiment of -120 poling cycles with gradual change of relative CEP by 2π was performed. Each poling cycle was the same as that of the first experiment. SH intensity change with time was plotted (Fig. 4). Numerical simulation using the model of the second experiment was performed taking the sample thickness into account. Sample thickness in the simulation was assumed to be $1.5 \mu\text{m}$. The simulation showed semi-quantitative agreement with the experiment (Fig. 4).

There are some notable differences from previous work[7] in the characteristics of the poling process because of the different conditions of the azo molecules. In the previous work, SCRP lead to linear decrease in SHG efficiency. This suggests that polarization created before SCRP coexists independently of the negative polar-

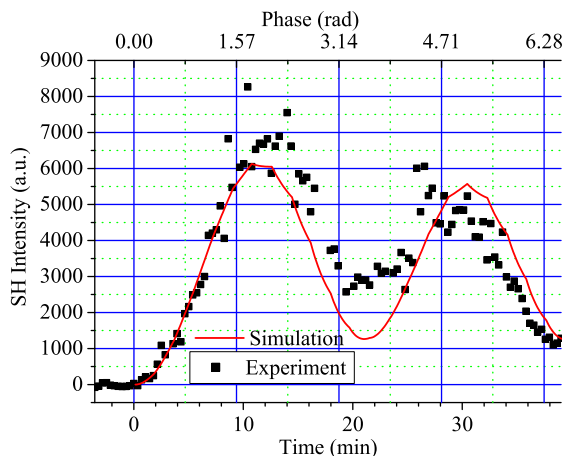


Fig. 4. SH intensity as a function of time during gradual carrier-envelope phase change reaching 2π . The line is obtained by numerical simulation using the model of the second experiment with phase mismatch due to the sample thickness taken into account.

ization created after SCRP. There is no polarization-restoring force in the previous case. On the other hand, in this work, the experimental results agree well with the model with polarization-restoring force. The origin of this difference is explained as follows. In the previous case, doped DR1 molecules had no bond to the polymer main chain and can change its orientation easily. In the present case, one end of DR1 molecules are attached to the main chain of the polymer with a covalent bond and molecular orientation change was restricted (Fig. 5). This difference illustrates the fact that the orientational behaviour of dye molecules in the polymer matrix is affected by the change of surrounding molecular environments.

An order of magnitude improvement in CEP detection speed achieved in this work suggests the possibility of further reduction of the time needed for CEP measurement. The polarization-restoring force found in this work might be used beneficially in such measurement. For example, in many applications of CEP measurement repetitive detection of CEP is required. If all-optical poling is used as the measurement process, some means to reset the generated polarization to zero is required. The simplest way to realize the reset is to move a sample stepwise to use a fresh part of the sample. However, inhomogeneity of density and thickness of the sample may induce problems. The presence of polarization-restoring force suggests that resetting to zero polarization can be achieved optically, eliminating the need of sample movement and associated problems.

Carrier-envelope phase measurement scheme presented in this work has the following advantages. Firstly, because the CEP signal is accumulated as material polarization in the sample, there is possibility of detecting weak signal with high signal to noise ratio by integration through simple increment of the poling time. Secondly, simple increase of beam power for the SHG efficiency measurement will lead to the quadratic increase of SH intensity hence higher signal to noise ratio. Although current SHG efficiency measurement was done using $1.6 \mu\text{m}$ light and erasure of induced polarization by 3-photon resonant absorption was observed, changing to longer wavelength will reduce 3-photon absorption and allow much higher beam power to be used to reduce the measuring time.

In conclusion, we showed that CEP can be determined by the all-optical poling of molecules with a large static dipole moment covalently bound to the polymer main chain. It was shown clearly the presence of polarization-restoring force in polar side-chain polymer which is absent in the previous work. This difference illustrates the fact that the rotation of the dye molecules in limited volumes is affected by the surrounding molecular environments. The polarization-restoring force is considered to be useful for resetting SH activity. Application of this method to repetitive CEP measurement might be feasible with further improvement in efficiency with even higher photochemical quantum efficiency and higher

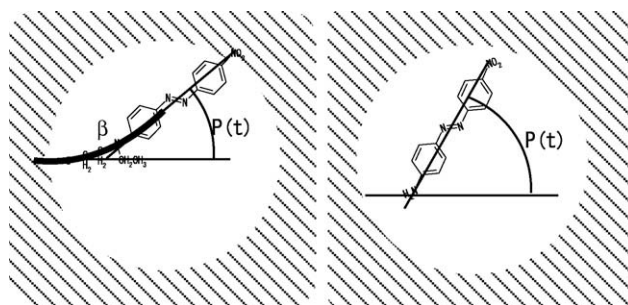


Fig. 5. Models to show the difference in molecular environment surrounding DR1 molecule. Left: poly(DR1 methacrylate). Right: DR1 doped in poly(methylmethacrylate).

concentration of molecules with larger dipole difference between the ground state and the excited state.

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