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# 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.



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 $\omega$ and $2\omega$ [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 $(2\omega)$ 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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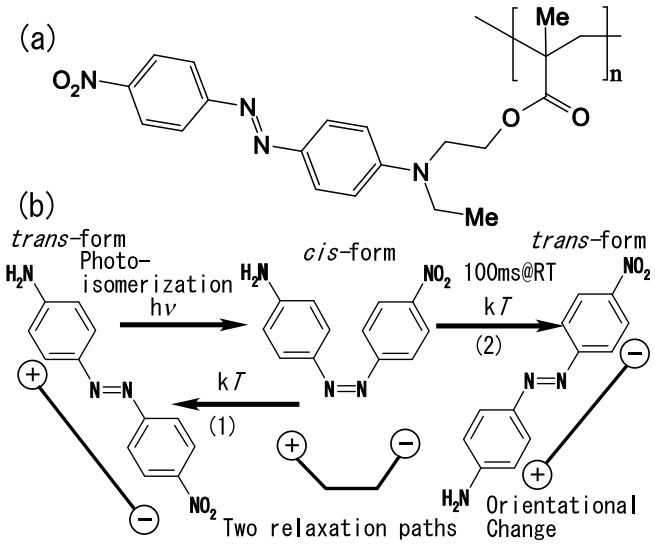


**Fig. 1.** (a) Poly(disperse red 1 methacrylate); (b) mechanism of all-optical poling utilizing *trans–cis–trans* photoisomerization cycle.

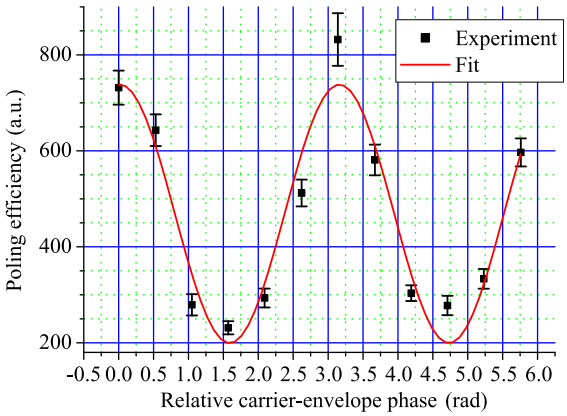


**Fig. 2.** All-optical poling efficiency as a function of relative carrier-envelope phase. Line shows a theoretical fit with $A + B\cos 2(\phi + \phi_0)$. $A = 470$, $B = 270$. The error bar of each point represents the standard deviation of parameter for each fitting of the SHG intensity in the each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

*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 $CaF_2$ 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–2f 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 $\omega$ light and the $2\omega$ 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 $CaF_2$ 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], 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 $\eta$. 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 $\cos(2(\phi_{\mathrm{rel}} - \phi_0))$ curve, in which $\phi_{\mathrm{rel}}$: relative CEP, $\phi_0$: initial CEP. The baseline offset from zero is due to the phase mismatch in finite thickness of the sample. Visibility $C$ defined as $C = (\eta_{\max} - \eta_{\min})/(\eta_{\max} + \eta_{\min})$ was $0.57 \pm 0.05$. Sample thickness estimated from optical density of the sample is 1.6 μm and visibility estimated from the sample thickness was $0.5 \pm 0.1$, 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 "SCRP" is used to designate the stepwise change of relative CEP by $\pi$ 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 SCRP in the middle of each run. A typical SHG intensity change plotted as a function of exposure time is shown in Fig. 3. Before SCRP, SHG intensity changes were the same as that of the first experiment and increased nearly linearly. Immediately after SCRP, 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 SCRP 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