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ОБЩАЯ И ТЕХНИЧЕСКАЯ ФИЗИКА

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PHOTOINDUCED SECOND ORDER SUSCEPTIBILITY BUILDUP STUDY BY USE OF SYNCHRONOUS WRITING AND PROBING TECHNIGUE

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Polymer optical materials become to be of increased interest because of their potential applications for electro–optic modulation, frequency conversion and data storage [1]. For example, high first hyperpolarizability of azo–dye molecules allows to obtain very large macroscopic $\chi^{(2)}$, if the molecules are aligned in proper way. Central symmetry of originally isotropic medium can be broken by applying of polar–asymmetrical optical field [2], for example, a mix of fundamental wave of frequency ω and coherent second harmonic (SH) with frequency 2ω (usually referred to as a "seeding" wave). This $\omega + 2\omega$ field has nonzero time average cube resulting in macroscopic spatially periodical $\chi^{(2)}$ ($\chi^{(2)}$ grating) proportional to $E_{\omega}^{2}(z)E_{2\omega}^{*}(z)\exp(-i\Delta kz)$, where z is a propagation coordinate, $\Delta k = k_{2\omega} - 2k_{\omega}$ is a wave vector mismatch. The process of creation of macroscopic $\chi^{(2)}$ susceptibility by polar–asymmetrical optical field is known as all–optical poling [3]. Optical poling method is of interest because of its simplicity and possibility to attain high $\chi^{(2)}$ under room temperature. This method can also serve as a mean for study of polymeric material nonlinear properties. However, photoinduced $\chi^{(2)}$ in polymeric materials is quasi–permanent and can be greatly influenced by probing radiation [4].

In this paper we present a new technique for real time monitoring $\chi^{(2)}$ during the poling process. This technique (referred to below as "real–time" method) can be used in a study of quasi–permanent photoinduced $\chi^{(2)}$ nonlinearity, because it excludes any unwanted meddling into poling process in contrast to the cases of alternating "writing and probing" [4] or using extra probe beam. Furthermore, our technique has been realized in the most simple and reliable configuration of co–propagating writing beams, since the same fundamental beam was used simultaneously as a pump and a probe.

Recent study of polarization properties of photoinduced $\chi^{(2)}$ in disperse red 1 (DR1) doped poly(methyl methacrylate) (PMMA) [5] has shown that the angle θ between planes of polarization of the photoinduced and seeding SH is nonzero in general case (i.e. when polarizations of the ω and 2ω beams are non–collinear). So, the seeding SH can be cut by an output polarizing analyzer, while the part of signal $I_{2\omega}^{signal}\sin^2\theta$ is transmitted through the analyzer and can be measured by a photodetector.

Experimental setup is shown in Fig. 1. Continuum Surelite–1 Q–switched Nd:YAG laser with 7 ns pulse width duration and 10 Hz repetition rate was used as a light source. The seeding SH was generated by a BBO crystal. Horizontal polarizer transmitted all fundamental radiation and small amount of SH. Quartz plates of 2 mm thickness were used to turn the polarization of seeding SH for an angle α different from 0^{o} or 90^{o} whereas the plane of fundamental beam polarization always remained horizontal. By turning two different quartz plates, the angle α could be turned to 37^{o} or 70^{o} respectively with satisfactorily linear ω and 2ω polarizations. The writing radiation was focused by a lens with 28–cm focal length. The energy of fundamental pulse was about 35 mJ, the energy of seeding light could be varied in wide range by the polarizer. The thickness of the sample was 1.4 μm and its optical density was 0.83. A photomultiplier tube (PMT) calibrated for 532 nm was used as a photodetector. The PMT response was always kept in linear range using calibrated neutral density filters. The signal from the PMT was integrated by a SRS Boxcar integrator and averaged over 200 shots by computer software. We compared SH growth curves obtained by our "real–time" and by traditional "writing and probing" techniques.

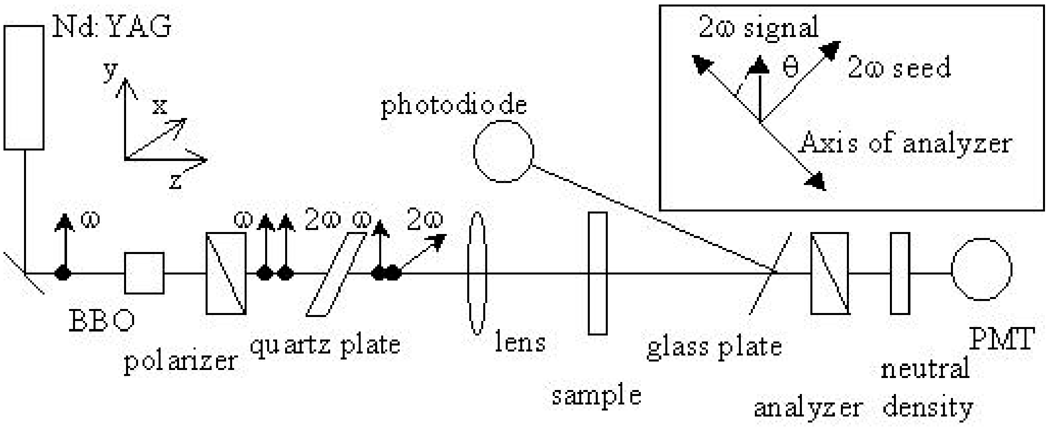

Fig. 1. Experimental setup for simultaneous writing and probing $\chi^{(2)}$ susceptibility in all–optical poling by co–propagating beams. The inset illustrates the basic principle of the method

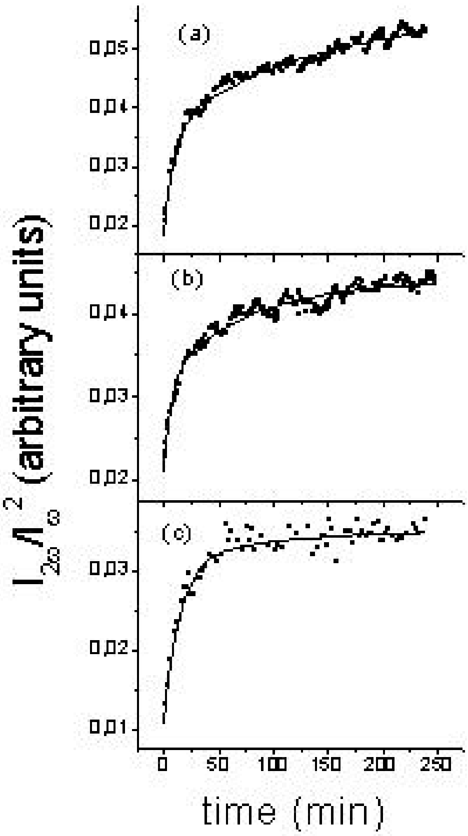

Fig. 2. Full normalized SH power detected after the analyzing polarizer as a function of all–optical poling time for:

a) "real–time" (α = 37°); b) "real–time" (α = 70°); c) "writing and probing" (α = 37°). Solid curves are the fits by a two–exponential function $f(t) = A + B_1(1 - \exp(-t/\tau_1)) + B_2(1 - \exp(-t/\tau_2))$

The SH growth curves are shown in Fig. 2. All the curves in Fig. 2 are fitted by a function $f(t) = A + B_1(1 - \exp(-t/\tau_1)) + B_2(1 - \exp(-t/\tau_2))$. This kind of "two–exponential" behavior can be explained on the base of simplified microscopic consideration showing two contributions to the SH signal: 1) short living conformation anisotropy, and 2) quasi–permanent net orientation emerging after reverse *cis–trans* transition [4].

Table 1

Fitting parameters in the function $f(t)=A+B_1(1- \exp(-t/\tau_1))+B_2(1- \exp(-t/\tau_2))$ obtained by successive approximations with Microcal Origin software

	τ_1, min	τ_2, min	B_1, a.u.	B_2, a.u.
Real–time ($\alpha = 37^o$)	7.72±0.41	122.3±4.8	0.01744±0.00043	0.01981±0.00022
Real–time ($\alpha = 70^o$)	7.16±0.54	93.6±3.0	0.01172±0.00038	0.01169±0.00013
Writing and probing ($\alpha = 37^o$)	13.87±0.92	84.9±25.5	0.01958±0.00101	0.00463±0.00047

The fitting parameters of the SH growth curves are summarized in Table 1. As illustrated in Table 1, there are several differences between the fitting parameters of "real–time" and "writing and probing" curves. First, τ_1 for "writing and probing" is about two times larger than that for "real–time" technique, and, second, the ratio of constants B_1 and B_2 is close to unity for "real–time", but about 4 for "writing and probing". It should be also noted that errors for the B_2 and τ_2 parameters is considerably larger in the case of "writing and probing", making difficult to identify the second mechanism. These differences seem to be due to destructive influence of reading radiation during 2 s long pauses in seeding. We have found that after few seconds of writing, the $\chi^{(2)}$ relaxes within about 5 seconds (see Fig. 3), which is comparable with the duration of reading (2 s). Every–minute interruptions also resulted in 1.5 times decrease of the maximum level of SH signal. Consequently, the influence of frequent meddling in $\chi^{(2)}$ building up cannot be neglected.

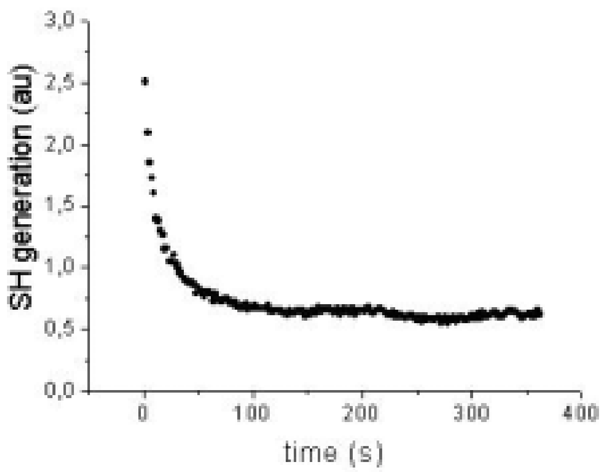

Fig. 3. Decay of the SH signal in the presence of the reading fundamental radiation

The $\chi^{(2)}$ was induced by simultaneous exposition to ω and 2ω during 3 seconds

The regular oscillations of the SH power especially noticeable in Fig. 2 (b) are associated with the SH generated in quartz plate. Since the SH from the quartz plate (about 1.5 pJ) was several orders of magnitude weaker than the seeding SH, it did not affect the real dynamics of $\chi^{(2)}$ in DR1–PMMA sample, but distorted the growth curve interfering with a part of seeding SH transmitted through the analyzer. This drawback can be eliminated by using a polarization rotator of different kind (e.g. Faraday's cell). The using of a SH free rotator can also increase the signal–noise ratio and make our technique equally applicable in rather wide range of angles between the writing beam polarizations (~30°—70°). Combining the real–time technique with the traditional "writing and probing" method [4], we checked that the curves in Fig. 2 represented the actual dynamic of $\chi^{(2)}$ in the thin–film sample.

Summary

In conclusion, we have demonstrated the real–time monitoring photoinduced $\chi^{(2)}$ during optical poling process using the same fundamental beam as a pump and a probe. We have shown that our technique can give certain advantage in comparison with more traditional “writing and probing” technique in dynamics studies of quasi–permanent photoinduced $\chi^{(2)}$. The method proposed can also facilitate all–optical poling process, because it does not need a computer controlled shutter. Our work on further development of the real–time technique and optimization of optical poling method is currently in progress.

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