Dynamic gratings of the quadratic polarizability in polydiacetylene PTS

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A single crystal of polydiacetylene PTS was exposed to $\langle \mathbf{E}^3 \rangle$-coherent picosecond pulses ($\lambda_\omega = 1.06\ \mu$m) and second-harmonic pulses ($\lambda_{2\omega} = 0.53\ \mu$m), which met at an angle $\theta = 4 \times 10^{-3}$ rad. The observed diffraction of the λ_ω beam through angles $-\theta$, -3θ, and -5θ is interpreted in terms of the writing of a grating of the quadratic polarizability and the subsequent generation of a $2\omega - \omega$ signal at this grating.

A field $\frac{1}{2}[E_\omega(\mathbf{r}) e^{-i\omega t} + E_{2\omega}(\mathbf{r}) e^{-i2\omega t} + \text{c.c}]$ has a polar asymmetry at a given point $\mathbf{r}$. This asymmetry is seen, in particular, in the circumstance that the cube of the field has a nonzero average value $\langle \mathbf{E}^3 \rangle = \frac{3}{8}(\mathrm{E}_\omega^2 \mathrm{E}_{2\omega}^* = \mathrm{E}_\omega^{*2} \mathrm{E}_{2\omega})$. Various effects result from the influence of such a field on a medium. For example, holographic gratings of the quadratic polarizability $\delta_\chi^{(2)} = \beta E_\omega^{*2}(\mathbf{r}) \mathrm{E}_{2\omega}(\mathbf{r}) + \text{c.c}$ can be written in fibers and glasses,[1] and a polar asymmetry occurs in the emission of electrons in the course of an interference of one- and two-photon ionization processes.[2]

In this letter we are reporting experiments on the effect of such fields on a single crystal of polydiacetylene PTS. The results indicate that a $\delta_\chi^{(2)} E_{2\omega} E_\omega^{*2} + \text{c.c}$ grating is written and that it is read out in the course of the generation of a signal at the difference frequency $2\omega - \omega$. The writing of $\delta_\chi^{(2)}$ gratings in the geometry of oppositely directed waves was studied experimentally in Ref. 3 for a solution of polydiacetylene 4BCMU.

In the experiments we used an Nd:YAG laser with passive mode locking. The length of an individual pulse in the train was about 50 ps. The divergence of the laser beam was 4×10^{-4} rad. The experimental arrangement is shown in Fig. 1. The light from the laser at the frequency ω was split in two by a beam splitter BS. The light which went through the optical delay line reached nonlinear-optics crystal *2*, in which a frequency doubling occurs. Glan prism *3* and the corresponding optical filter made

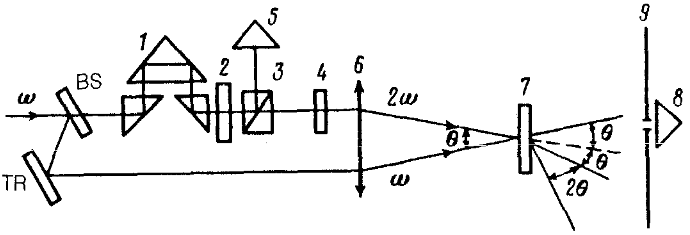

FIG. 1. Experimental layout. *1*—Optical delay; *2*—KTP nonlinear-optics crystal; *3*—Glan prism; *4*—SZS-23 filter; *5*—photodiode; *6*—lens ($f=140$ cm); *7*—the test sample, a crystal of polydiacetylene PTS; *8*—photodiode; *9*—diaphragm.

it possible to select the signal at the frequency 2ω alone, with a polarization vector parallel to the X axis. The divergence of the light at the second harmonic did not exceed 4×10^{-4} rad. The ω pulse reflected from BS was reflected once more by "total"-reflection mirror TR and was directed, along with the pulse at 2ω, to the polydiacetylene PTS sample by lens 6 (the focal length of this lens was 140 cm). The diameter of the ω and 2ω beams at the entrance to the lens did not exceed 1.5 mm. The angle at which these two beams met was $\theta_m=4\times10^{-3}$ rad. The polarization vector of each beam was parallel to the polymerization filaments in the PTS crystal.

To measure the intensity of the diffracted light at the frequency ω in the far zone, we used photodiode *8*, which we moved in the plane in which the beam met. This photodiode had a slit diaphragm *9* with a width of 200 μm. The distance from the sample to the photodiode was 150 cm. Photodiode *5* was used to monitor the incident light.

Figure 2 shows the experimental angular distribution of the intensity of the diffracted light at the frequency ω. In measuring this dependence we monitored the diffraction at 2ω by replacing the interference filter by photodiode *8*. We should point out immediately that we did not observe a diffraction at the frequency 2ω at any other angle, except for a weak zeroth-order scattering. The PTS polymer crystal has an absorption coefficient[4] $\sigma=2\times10^{+5}$ cm^{-1} at the wavelength of the second harmonic of a neodymium laser. Accordingly, if any scattering does occur at the frequency 2ω, it will be completely absorbed inside the crystal. The absorption at the frequency ω was less than 1%.

To explain the experimental behavior found, we consider three mechanisms for the formation of diffracted waves at the angle $-\theta$, -3θ, and -5θ (Fig. 2).

As light at the frequency ω, i.e., E^0_ω, penetrates into the crystal, it may generate the second harmonic, by (for example) a quadrupole mechanism. In the course of an interaction with the incident second harmonic, $\mathrm{E}^0_{2\omega}$, both types of light may lead to the formation of a grating of the dielectric constant, $\delta\epsilon$, with a wave vector $\mathbf{q}=\mathbf{k}_{2\omega}-2\mathbf{k}_\omega$. The incident light at both ω and 2ω is diffracted by this grating. The latter frequency could not be observed in our case, for the reason stated above. The diffraction at the frequency ω is a Raman–Nath diffraction, specifically, at angles $\pm2\theta$, $\pm4\theta$, ... with respect to the direction of the incident light E^0_ω. The intensity of the diffracted light at

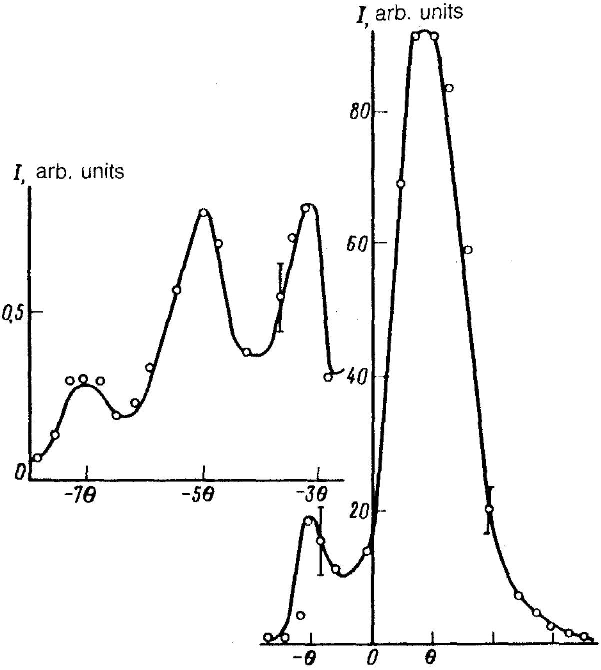

FIG. 2. Angular distribution of the light intensity at the frequency ω in the far zone.

the $+1$ diffraction maximum, i.e., I_ω^{+1}, depends on the intensity (I_ω^0) of the incident light at the frequency ω in accordance with $I_\omega^{+1} \propto (I_\omega^0)^3$.

Another likely mechanism for the appearance of diffracted waves might be as follows: When a sample is illuminated by coherent waves at ω and 2ω simultaneously, a grating of the quadratic nonlinear polarizability, $\delta_\chi^{(2)} = \beta E_{2\omega} E_\omega^{*2} \exp(i\mathbf{qr}) + \text{c.c.}$, may form, where $\mathbf{q} = \mathbf{k}_{2\omega} - 2\mathbf{k}_\omega$ is the grating vector. The difference frequency $E_{2\omega} E_\omega^*$ is generated at this grating. It is not difficult to show that the scattering at the resonant frequency occurs along the angle -4θ with respect to E_ω^0, which coincides with the -3θ direction in Fig. 2. Since we have $n_\omega > n_{2\omega}$, the grating vector is tilted in such a way that the mismatch of the wave vector is smaller along -3θ than along $+\theta$. The scattering by the grating vector $2\mathbf{q}$ results in the appearance of a wave in the -5θ direction. The mismatch of the wave vector is even smaller. Along $+3\theta$, in contrast, the mismatch increases; as a result, there is pronounced weakening of the scattered wave. This mechanism does not explain the appearance of a wave at ω in the $-\theta$ direction. On the other hand, the intensity of the scattered signal in this direction was a linear function of the intensity I_ω^0. For this reason, the first mechanism also fails to explain this result.

The difference frequency $E_{2\omega} E_\omega^*$ may be generated in the $-\theta$ direction by a quadrupole mechanism. In this case the intensity of the signal would be a linear function of the intensity of the input signal at ω.

In summary, a wave of the difference frequency $E_{2\omega} E_\omega^*$ is generated in the $-\theta$ direction at the front face of a PTS crystal (Fig. 2). The simultaneous illumination of the crystal with an electromagnetic field with $\langle \mathbf{E}^3 \rangle = 0$ gives rise to a $\delta_{\chi}^{(2)}$ grating, which scatters the difference frequency. The generation of a difference frequency at the angles $+\theta$, -3θ, -5θ, ... can occur at the same grating. The scattering into positive angles is less likely than the scattering into negative angles. We should point out that in these experiments we were unable to observe any scattering at the frequency ω at observation angles greater than θ.

A dynamic grating of the quadratic polarization has thus been observed experimentally in a PTS crystal. The nature of this grating is unclear at this point.

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