Laser-induced holographic light scattering in a liquid-crystalline azobenzene-containing polymer

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The holographic scattering phenomenon was observed and studied in detail for azobenzene-containing polymer system. It was found that irradiation of 25-μm-thick samples of the polymer with a single linearly polarized laser beam results in the appearance of a band with the maximum in the vicinity of irradiating wavelength in polarized extinction spectra. The band is completely polarized along the polarization direction of irradiating light. The shape of the band is exceedingly sensitive to rotation of the sample with respect to the spectrometer probe beam. An analogous band appears upon the irradiation of the sample with circularly polarized light. The observed phenomenon is presumably similar to holographic scattering, reported previously in photorefractive crystals and some photopolymers. A detailed description of the observed spectral changes is presented. The sensitivity of the effect towards probe light polarization and direction, irradiating light wavelength, and temperature is discussed.

I. INTRODUCTION

Amorphous and liquid-crystalline polymers containing photoactive azobenzene moieties comprise a promising class of photoactive media [1,2]. These materials are photoanisotropic because the irradiation with linearly polarized light induces an orientation of azobenzene chromophores perpendicular to the electric-field vector of light. This process is referred to as photo-orientation [3–13]. The orientation process is cooperative, i.e., the alignment of azobenzene moieties is accompanied by the alignment of nonphotoactive molecular fragments [3,4,8,9]. The mechanism of cooperative alignment has been discussed in the literature [14–19]. Photoorientation results in high photoinduced macroscopic birefringence and dichroism in irradiated materials, which allows one to use these systems as holographic media. The inscription of both thin and volume holograms in azobenzene-containing polymer films irradiated with two laser beams has been extensively studied by various authors [8,20–24].

Upon the irradiation of a thick sample of azobenzene-containing liquid-crystalline polymer with a single linearly polarized laser beam, we came across an unexpected phenomenon, manifested in unusual changes of the UV-visible extinction spectrum of the sample during irradiation. We believe that these changes are associated with the phenomenon known in the literature as holographic scattering.

Holographic scattering has been known for several decades [25]. Its physical basis is the interference of irradiating beam with light scattered by defects within or on the surface of the irradiated sample. The resulting interference pattern is inscribed within the photosensitive medium as a set of holographic gratings. These gratings in turn act upon the irradiating beam to result in enhanced scattering, thus providing positive feedback for the inscription process. This mechanism leads to a significant increase of scattering of the sample during its irradiation. Holographic scattering has been observed in some photoactive materials, namely, photorefractive crystals [26–29], photosensitized polymethyl methacrylate [30,31], acrylamide photopolymer [32], polymer dispersed liquid crystals [33], and silver halide photoemulsions [34–37]. This phenomenon, holographic in its nature, occurs upon the irradiation of the sample with a single laser beam, as opposed to a conventional double-beam holographic recording.

Although light scattering has been shown to impair the quality of holograms in liquid-crystalline azobenzene-containing polymers [38], holographic scattering similar to observed in photorefractive crystals and photopolymers [25–37], to our knowledge, has not been reported in azobenzene-containing materials.

In this paper we describe an unexpected laser-induced phenomenon in azobenzene-containing polymer. We intend to describe the details of the phenomenon observed and present our arguments in favor of its holographic scattering nature.

II. EXPERIMENTAL DETAILS

We investigate the azobenzene-containing polymer PAAzo6, the structure of which is presented in Fig. 1(a). The synthesis of the polymer is described elsewhere [39]. Transition temperatures for the polymer are $T_g = 24\,°C$ (glassy to nematic), $T_{NI} = 120\,°C$ (nematic to isotropic). The vast temperature interval of the nematic phase is favored by the presence of phenylbenzoate nematogenic fragments in the polymer structure.

It is known [39,40] that thin films of the polymer studied exhibit photo-orientation upon irradiation with linearly polarized visible light [Fig. 1(b)]. The preferential orientation of chromophores perpendicular to irradiating light polarization direction is observed.

For the sample preparation polymer melt at $120\,°C$ was pressed between two glass plates and then cooled to ambient temperature with the rate of approximately $20\,°C/min$. The film thickness was regulated by glass spacers, 25 or 11 μm thick, placed between the glass plates. Before each experiment the polymer sample was annealed at $50\,°C$ for 2 h.

The samples were irradiated with frequency-doubled diode-pumped YAG lasers (532 nm, 12 mW/cm^2 and 473 nm, 20 mW/cm^2) and the laser beam was directed normally on the sample surface. The intensity of irradiating light was controlled with a Vishay BPW21R photodiode. The laser beam was expanded so that the whole sample area was irradiated. To increase the degree of light polarization, the irradiating beam was passed through a Glan-Taylor prism. Circularly polarized light was obtained with a Hoya CIR-PL circular polarizer.
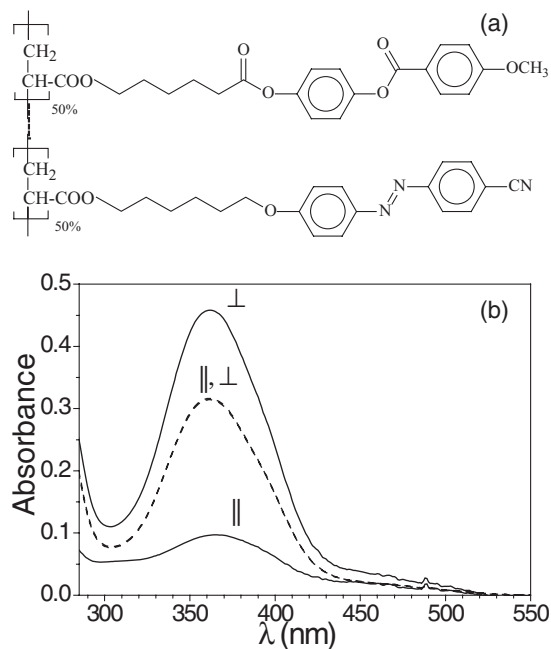
FIG. 1. (a) Structure of polymer PAAzo6 and (b) the UV-visible spectrum of a 300-nm-thick PAAzo6 film before (dashed line) and after (solid line) irradiation with 473-nm linearly polarized laser light.

Light extinction was measured with a UV-visible spectrophotometer Specord M-40. The light collection angle estimated on the basis of the detecting system geometry was approximately $13°$. In this work the extinction is presented as $\log_{10}(I_0/I)$ [optical density (OD)], where I_0 and I are the intensities of incident and transmitted light, respectively. In order to record optical spectra in polarized light, an appropriate polarizer was placed in the path of the spectrometer probe beam before the sample.

Scattering measurements were performed using the setup sketched in Fig. 2. A focused laser beam 1, after passing through the Glan-Taylor prism 2 and iris diaphragm 3, fell on the sample 4. The intensity of scattered light was measured with a photodiode detector mounted on a 27-cm-long mobile arm. The arm, controlled by a stepping motor, could be rotated around the sample by an arbitrary angle α. To control light intensity, the probe beam was partially reflected by a glass plate 7 and the reflected intensity was measured with a photodiode 8. The angular resolution of the setup was $1°$.

The obtained scattering indicatrices were corrected to account for light refraction. The scattered light, propagating within the polymer by an angle β to the sample normal, refracts
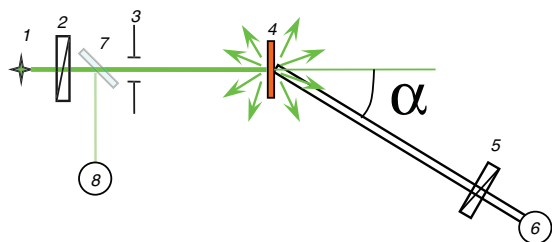

FIG. 2. (Color online) Experimental setup for scattering measurements (see the text).

at polymer-glass and glass-air interfaces and propagates in the air by an angle α to the sample normal. The relation between α and β is given by Snell's law $\sin\alpha = n_{\text{polymer}}\sin\beta$, where n_{polymer} is the average refractive index of the polymer film. The value of refractive index was taken to be 1.6 on the basis of measurements for similar compounds [41]. The light scattered by angles β in the interval

$$\beta \in \left[\arcsin\left(\frac{1}{n_{\text{polymer}}}\right); 180° - \arcsin\left(\frac{1}{n_{\text{polymer}}}\right)\right]$$
$$\approx (39°; 141°)$$

undergoes total internal reflection and therefore the intensity of light scattered in these directions cannot be measured using the described apparatus. In the experimental indicatrices presented in Sec. III the scattering angles correspond to light propagation within the polymer (β angles).

III. RESULTS AND DISCUSSION

Upon the irradiation of the polymer with linearly polarized laser light ($\lambda = 532$ nm) the following changes in polarized UV-visible extinction spectra are observed (Fig. 3). In the first minutes of irradiation, the extinction in the shorter-wavelength part of the spectrum increases in both polarizations [Fig. 3(b), inset]. This change can be attributed to trans-cis isomerization of azobenzene moieties towards the photostationary state [2, 42,43]. Upon further irradiation the scattering of the sample increases and liquid-crystalline domains become finer, as seen in polarization microscope images [Figs. 4(a) and 4(b)]. Isomerization completes within a few minutes, whereas the increase of extinction due to scattering takes tens of minutes to hours.

Concurrently with the increase of scattering, an unexpected phenomenon occurs. A band, centered in the vicinity of the irradiating wavelength, appears in the spectra [Fig. 3(a)]. This band is completely polarized parallel to the irradiating light polarization. In the experiment illustrated in Fig. 3, the photostationary state is reached in approximately 250 min, which corresponds to 180 J/cm^2 light energy. The irradiation with noncoherent light (mercury lamp, 546 nm) of the same intensity does not result into the appearance of the band described above.

The shape of the laser-induced band is notably sensitive to the angle of incidence of the spectrophotometer probe beam. Rotation of the sample in the spectrophotometer [Fig. 5(a)] leads to doubling [rotation around the axis perpendicular to irradiating light polarization, Fig. 5(b)] or widening [rotation around the axis parallel to irradiating light polarization, Fig. 5(c)] of the band. Turning the sample by small angles results in significant changes in the spectrum shape. To accentuate the change of the laser-induced band shape and level the intrinsic absorption of azobenzene moieties, the difference spectra between parallel and perpendicular extinction are presented in Figs. 5(b) and 5(c).

The described phenomena, i.e., the appearance of a laser-induced band in the vicinity of the irradiating wavelength and the change of the spectrum shape with the sample rotation, are strongly polarization dependent and are not observed in the spectra polarized orthogonally with respect to the irradiating light.
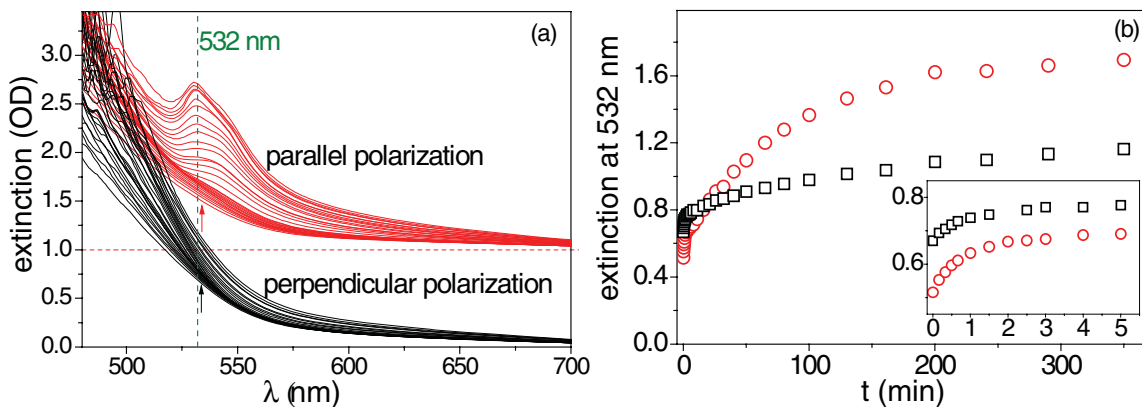
FIG. 3. (Color online) (a) Change in polarized UV-visible extinction spectra during irradiation with polarized laser light at 532 nm (spectra with parallel polarization are shifted up by 1.0 to increase legibility) and (b) kinetics of extinction change at the irradiating wavelength (the inset depicts the initial course of the kinetic curve). Polarization of the probe light is parallel (circles) or perpendicular (squares) to the polarization of the irradiating light.

When the same polymer was irradiated with 473-nm laser light, an analogous band in the spectrum (with the maximum at 478 nm) appears (Fig. 6).

The described phenomenon is found to be reversible. Annealing the sample in the nematic phase leads to the erasure of the laser-induced band. The rate of the erasure process is temperature dependent; it takes approximately 10 min at 40 °C and approximately 100 min at 35 °C. Annealing at 40–50 °C for several hours leads to the enlargement of liquid-crystalline domains and the decrease of light scattering.

The band in the vicinity of the irradiating wavelength also appears upon the irradiation of the sample with circularly polarized laser light. The laser-induced band in this case is not seen when probing with orthogonally circularly polarized light (Fig. 7).

Thus, the following are the main features of the described photoinduced spectral band.

(i) The band appears only upon irradiation with coherent light.

(ii) The band is completely polarized; its polarization is identical to the polarization of irradiating light (i.e., the band has the highest intensity when probing with light of the same polarization and disappears when probing by orthogonally polarized light).

(iii) The band shape is sensitive to the orientation of the probe beam with respect to the sample normal.
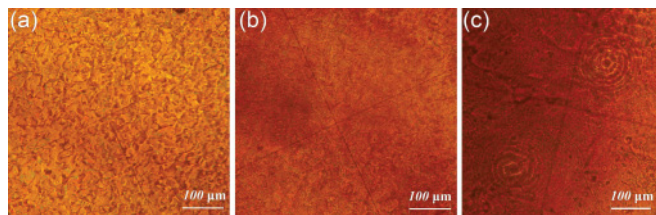

FIG. 4. (Color online) Polarization microscope images of the sample (a) before irradiation, (b) after 30 min of irradiation with 532-nm linearly polarized laser light, and (c) after 300 min of irradiation with 532-nm linearly polarized focused laser beam.
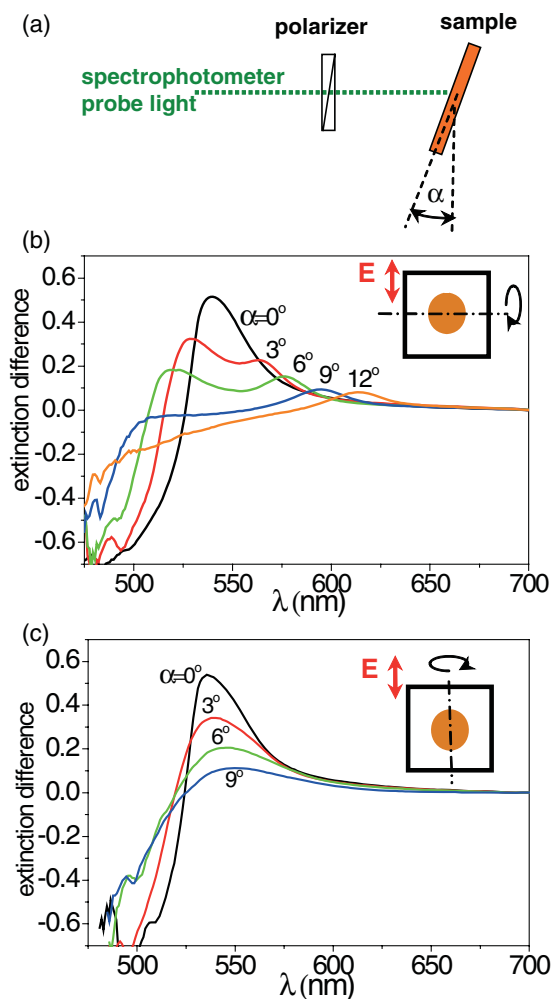

FIG. 5. (Color online) Change of UV-visible spectra of the irradiated sample of PAAzo6 on turning the sample with respect to the spectrometer probe light: (a) scheme of the experiment. Also shown is the change of the difference spectra observed when rotating the sample around the axis (b) perpendicular and (c) parallel to irradiating light polarization.
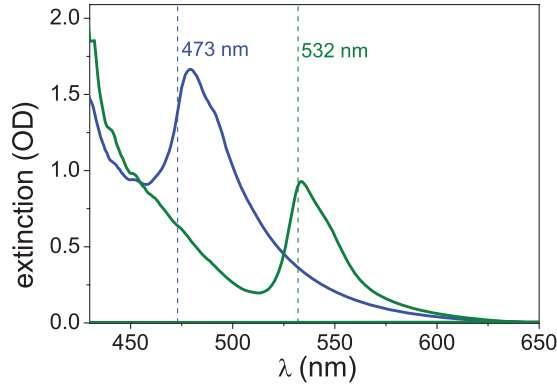
FIG. 6. (Color online) Spectra of the samples irradiated with 473- and 532-nm laser light. The film thickness is 11 μm.
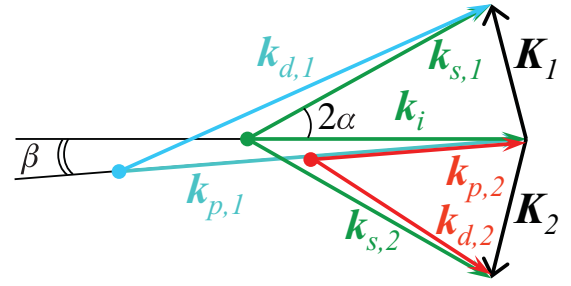

FIG. 8. (Color online) Sketch explaining the splitting of the selective reflection band on rotating the sample with respect to the probe beam (see the text).

The peculiarities listed above indicate that the observed band in the UV-visible spectra is not caused by mere light absorption, but by a more complicated optical phenomenon. The dependence of the band maximum on the irradiating wavelength and sensitivity toward sample rotation indicate that a Bragg diffraction grating with the grating period determined by the irradiating wavelength appears in the polymer upon laser irradiation. We believe that this diffraction grating appears as a result of the interference of the irradiating beam with the light coherently scattered within the sample. The interference pattern is holographically inscribed within the material due to photo-orientation of the polymer sidegroups in the direction perpendicular to light polarization [1,2,13]. This phenomenon has been described in the literature and is usually referred to as holographic scattering [25–37]. In the present paper we report on holographic scattering in an azobenzene-containing material.

The holographic scattering mechanism is confirmed by the splitting of the band in the spectrum on turning the sample, as seen in Fig. 5(b). This phenomenon can be qualitatively explained as follows. Let us consider the interaction of the irradiating beam with wave vector $\mathbf{k_i}$ with beams, symmetrically scattered by angles 2α and -2α and having wave vectors $\mathbf{k_{s,1}}$ and $\mathbf{k_{s,2}}$ (Fig. 8). Interference of the irradiating light with the

scattered light and holographic inscription of the interference pattern leads to formation of two gratings with vectors

$$\begin{aligned} \mathbf{K_1} &= \mathbf{k_{s,1}} - \mathbf{k_i}, \\ \mathbf{K_2} &= \mathbf{k_{s,2}} - \mathbf{k_i}, \end{aligned} \tag{1}$$

which are symmetric about $\mathbf{k_i}$. If the probe beam coincides with the irradiating beam, then the Bragg condition for both gratings $\mathbf{K_1}$ and $\mathbf{K_2}$ is automatically fulfilled at the irradiating wavelength and a selective reflection band at this wavelength should appear in the transmittance spectrum. Let us now suppose that the probe beam is directed at an angle β with respect to the irradiating beam. In this case $\mathbf{K_1}$ and $\mathbf{K_2}$ are no longer symmetric about the probe beam and the Bragg condition for them is fulfilled at different wavelengths. Therefore, on turning of the sample with respect to the probe beam, the line in the spectrum, corresponding to selective reflection on gratings $\mathbf{K_1}$ and $\mathbf{K_2}$, should split in two. In Fig. 8, $\mathbf{k_{p,1}}$ and $\mathbf{k_{p,2}}$, correspond to probe light wave vectors, which meet the Bragg condition for gratings $\mathbf{K_1}$ and $\mathbf{K_2}$, and vectors $\mathbf{k_{d,1}}$ and $\mathbf{k_{d,2}}$ correspond to selectively reflected light. From the Bragg conditions

$$\begin{aligned} |\mathbf{k_{d,1}}| &\equiv |\mathbf{k_{p,1}} + \mathbf{K_1}| = |\mathbf{k_{p,1}}|, \\ |\mathbf{k_{d,2}}| &\equiv |\mathbf{k_{p,2}} + \mathbf{K_2}| = |\mathbf{k_{p,2}}| \end{aligned} \tag{2}$$
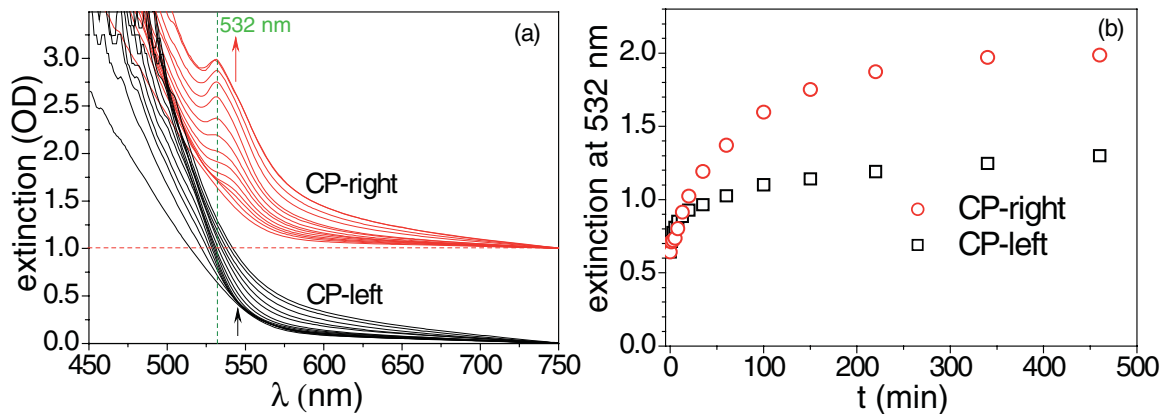

FIG. 7. (Color online) (a) Change in UV-visible extinction spectra recorded in left (CP-left) and right (CP-right) circularly polarized light during irradiation with RCP laser light at 532 nm (CP-right spectra are shifted up by 1.0 to increase legibility) and (b) kinetics of the extinction change at the irradiating wavelength.

and geometric considerations, selective reflection wavelengths are given by

$$
\lambda_{Br,1} = \frac{n_i}{n_p} \lambda_0 \frac{\sin(\alpha - \beta)}{\sin \alpha},
$$
$$
\lambda_{Br,2} = \frac{n_i}{n_p} \lambda_0 \frac{\sin(\alpha + \beta)}{\sin \alpha}, \quad (3)
$$

where $\lambda_{Br,1}$ and $\lambda_{Br,2}$ are Bragg wavelengths for gratings $\mathbf{K_1}$ and $\mathbf{K_2}$ and n_i and n_p are refractive indices for irradiating and probe light, correspondingly.

Conventionally, the experimental angular dependences of holographic scattering are recorded at a constant wavelength and varying angle. For this experimental configuration the above-stated consideration predicts two symmetric minima in the dependence of the transmittance on the sample rotation angle. These two minima correspond to Bragg angles $\pm\beta_{Br}$, which satisfy Eq. (3) at a constant probe light wavelength. Such an angular dependence with two minima has been observed in holographic scattering experiments in various systems and is admitted to be a characteristic feature of holographic scattering [25,28,29,34]. The simple model presented here accords with theoretical considerations found in the literature for holographic scattering [28].

The presented model, aimed to qualitatively explain spectral band splitting, is oversimplified for several reasons. First, it considers only a two-dimensional case, when the scattering occurs in the plane perpendicular to sample rotation axis, and therefore wave vectors of the irradiating light, scattered light, and probe light lie in the same plane. Second, the light is not scattered in a single preferential direction, but generally in all directions. Therefore, the observed structure is a superposition of many gratings. The diffraction efficiency of pairs of gratings $\mathbf{K_{1,2}}(\alpha)$ is determined by the intensity of light scattered to the angle 2α. When the sample is turned with respect to the probe light, the spectrum components corresponding to different grating pairs should split differently, depending on α. Different displacements of different spectrum components should lead to broadening of the overall spectrum on the sample rotation, which is indeed observed in our experiments [Fig. 5(b) and 5(c)].

As shown in Fig. 6, in our experiments the laser-induced band is slightly shifted from the irradiating wavelength. This deviation can be explained by slightly inorthogonal mounting of the sample in the spectrophotometer or by thermal dilatation of the polymer in the course of irradiation.

Corroborating the holographic scattering mechanism on the described observation is the appearance of characteristic texture elements on the irradiated sample surface after long irradiation [Fig. 4(c)]. These structures, having the shape of concentric rings, presumably appear as a result of the holographic inscription of light scattered on large inhomogeneities of the sample material. They are not observed on nonirradiated areas of the sample surface. Unlike holographic gratings from domain boundaries, defects, and other small scattering centers, holographic gratings from light scattered on large inhomogeneities can be observed directly and therefore comprise a demonstrative special case of holographic scattering. For the observation of the concentric rings, the microscope must be focused on the very surface of the film, implying that the
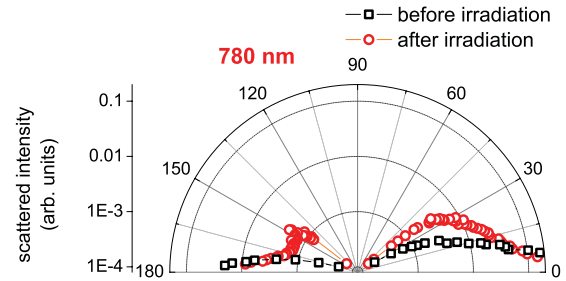

FIG. 9. (Color online) Scattering indicatrices of 780-nm laser light before (squares) and after (circles) 532-nm laser irradiation.

corresponding structures are localized in the very vicinity of the irradiated surface.

To elucidate the details of holographic scattering in the studied azobenzene-containing polymer, scattering indicatrices of the sample were experimentally measured using the setup presented in Fig. 2. Probe light with wavelengths of 532 and 780 nm was used. The polarization of scattered light could be measured in our experiments.

Experimental scattering indicatrices of infrared laser light (780 nm) *before* and *after* irradiation with a 532-nm laser are presented in Fig. 9. These indicatrices do not depend on the polarization of probe light with respect to the irradiating light. The scattered light was found to be nonpolarized. It is evident from Fig. 9 that the irradiation results in a significant increase of light scattering (note the logarithmic scale of intensities).

The increase of scattering during irradiation is illustrated in Fig. 10, where the time dependence of the light intensity, scattered by 160°, is presented. The sample was irradiated with a vertically polarized focused laser beam at a wavelength of 532 nm.

In the photostationary state (after long irradiation with a 532-nm laser) the scattering indicatrices were recorded at the irradiating wavelength. The experimental results are presented in Fig. 11. One could see that the probe light is scattered differently, depending on its polarization. The peculiar case is when the probe and the irradiating beams are of the same polarization (Fig. 11, squares). A great amount of backscattering (diffuse reflection) is observed in this case. It is specific to scattering of the probe light of the same wavelength and polarization as the irradiating beam and is not
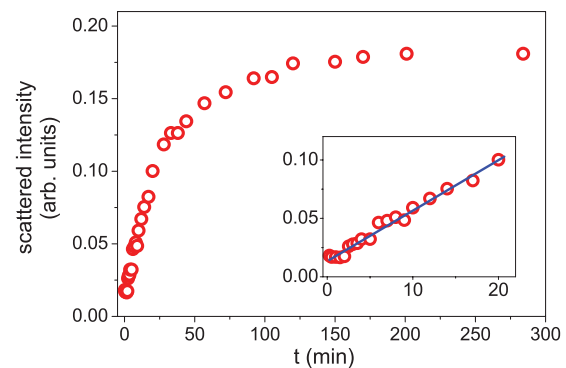

FIG. 10. (Color online) Kinetics of the change of intensity scattered by 160° during irradiation of the sample with 532-nm laser light (the inset illustrates the initial course of the kinetics). The irradiating light (532 nm) serves as the probe light.
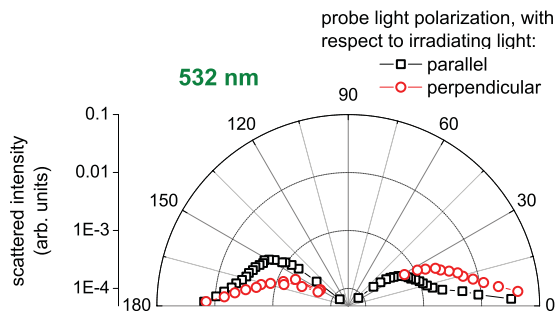
FIG. 11. (Color online) Dependence of light scattering indicatrices on the probe light polarization in the photostationary state. The probe light polarization is parallel (squares) and orthogonal (circles) to the irradiating light polarization.
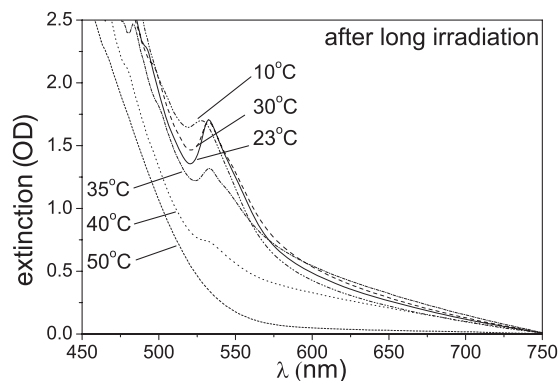

FIG. 12. Ultraviolet-visible extinction spectra of the PAAzo sample after prolonged irradiation with 532-nm laser light at 10–70 °C. The polarization of the probe light is coincident with the polarization of the irradiating light.

observed when probing with orthogonally polarized 532-nm light (Fig. 11, circles) or 780-nm light (Fig. 9, circles). In the experiment, illustrated in Fig. 11, the scattering was measured in the plane normal to the irradiating light polarization direction. In the case of scattering in the plane parallel to irradiating light polarization, the picture is qualitatively the same, although the sensitivity to probe light polarization is less pronounced.

The measurements of the polarization of the scattered light show that the forward-scattered light is nonpolarized, which can be explained by depolarization within the polydomain liquid-crystalline sample, whereas the diffusely reflected light is polarized the same way as the probe beam. This implies that the irradiated sample consists of two optically dissimilar layers. In the vicinity of the irradiated surface the holographic structure effectively diffracts light of the same polarization as the irradiating light; however, the rest of the film is composed of polydomain liquid-crystalline polymer, which depolarizes light.

From the measurements of scattering indicatrices the following conclusions can be drawn.

(i) In the course of laser irradiation, the scattering significantly increases.

(ii) The scattering of light at the irradiating wavelength is sensitive to its polarization: The probe light polarized as the irradiating beam is selectively backscattered, whereas orthogonally polarized light is scattered by small angles.

(iii) The backscattered light conserves its polarization. The light scattered forward is depolarized.

Thus, both extinction spectra and scattering measurements prove the marked sensitivity of holographic scattering toward the probe light polarization. Selective backscattering and a Bragg reflection band appear only in the light of the same polarization as that of the irradiating light. Exceeding polarization sensitivity is a specific feature of holographic scattering in the studied azobenzene-containing polymer. As it was shown in Refs. [29,36,37] for photorefractive crystals and silver halide photoemulsions, the intensity of holographic scattering is somewhat reduced when probing with orthogonally polarized light. The effect is explained by scattering anisotropy [36,37] and different refractive indices for different polarizations [29]. In our case, however, the effect of probe light polarization is much more pronounced. Apparently, it is caused by the polarization holographic mechanism of holographic recording in the

studied material. It is well known that polarization holograms inscribed in azobenzene-containing polymers can have zero efficiency at an orthogonally polarized reading [44,45].

Finally, temperature dependence of holographic scattering efficiency was investigated. In Fig. 12 photostationary parallel UV-visible spectra after long irradiation at various temperatures are presented. The highest efficiency of holographic scattering is reached at 23 °C, which is close to the glass transition point of the studied polymer ($T_g = 24$ °C). The irradiation of the sample at lower or higher temperatures leads to the Bragg band of lower intensity. At lower temperatures the holographic inscription may be impeded by the hindered rotational mobility of azobenzene molecular fragments, whereas at temperatures higher than T_g the disruption of photoinduced order occurs. Thus, in the vicinity of T_g, the balance is kept between the effective inscription and the stability of the scattering hologram.

It is noteworthy that the appearance of a reflection band in a circular dichroism spectrum upon the irradiation of an azobenzene-containing polymer with circularly polarized light has been previously reported in Ref. [46]. This band was centered at the irradiation wavelength. Upon the sample rotation the band changed its shape in a way similar to that shown in Fig. 5(b). The authors explained their observations by a supramolecular helical structure induced in the sample by a combination of the circularly polarized irradiating beam with the light reflected back from a glass substrate of the sample. However, numerical simulations [47] showed that this mechanism is unlikely to be consistent with the sharp Bragg band observed experimentally. Perhaps the observations described in Ref. [46] can be alternatively explained in the framework of the holographic scattering model proposed in the present paper.

IV. CONCLUSION

In the present paper, the phenomenon of holographic light scattering induced in an azobenzene-containing polymer upon irradiation with a single laser beam has been described. The kinetics of the inscription process and temperature stability of laser-induced structures is described. The dependence of the sample transmittance on the probe light polarization and

sample orientation has been determined experimentally. The main peculiarity of holographic scattering in the studied azobenzene-containing polymer is an exceptional sensitivity of the effect to probe light polarization, which is presumably associated with the polarization holographic nature of photoinduced scattering.

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