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Temporal evolution of photoinduced optical chirality in nanostructured light-sensitive waveguide thin films: Simultaneous excitation of TE$_0$ and TE$_1$ modes

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ABSTRACT
Optical properties of the self-organized gratings in thin photosensitive waveguide AgCl films on a glass substrate, doped by Ag nanoparticles, in the case of simultaneous excitation of TE$_0$ and TE$_1$ modes are investigated. These gratings are a result of interference of the incident linear polarized He-Ne laser beam with the excited TE$_n$-modes of the AgCl layer. The interference field patterns the re-distribution of the nanoparticles. Each excited mode generates its own grating. The self-organized gratings associated with the TE$_0$ mode appear earlier than that of the TE$_1$ mode. There is a competition between TE$_0$-gratings and TE$_1$-gratings for absorbing the nanoparticles, because their available amount is limited in the interaction area. The competition is due to the positive feedback of the light mechanism. Thus, the TE$_1$-grating removes some Ag nanoparticles from the already generated TE$_0$-grating, which forms some chiral complex building blocks of the gratings. The induced chirality leads to the induction and enhancement of optical activities in the samples. Longer exposure of the samples leads to the variation of induced optical chirality. Generation of the gratings increases anisotropy of the sample, which in turn enhances the chiroptical properties of the Ag/AgCl/Glass system. Changes in absorption spectra, induced dichroism, and induced optical gyrotropy of the samples are measured. Diffraction patterns of the exposed samples confirm the coexistence of TE$_0$- and TE$_1$-associated gratings and their competition. AFM images support the above-mentioned process. It is observed that longer exposure and perfect formation of the induced gratings accompany with the reduction of chiroptical property, due to the destruction of chiral complex silver nanoclusters.

I. INTRODUCTION
Formation of self-organized periodic nanostructures (SPNs) in light-sensitive planar waveguide thin films, as a result of the interaction with a low power laser beam, has been studied in detail for many years. Since this system is very simple and at the same time is capable of recording information about polarization (its sign of rotation and state), the wavelength of the incident light, and even the index of refraction of the substrate, to improve the process, it would be important to find ways to make this optical nano-structuring method faster and more efficient. On the other hand, silver is a very commonly available material. Its nanoparticle preparation is relatively easy. Besides that, it gives photosensitivity to Ag-Hal (Hal for halides) thin films, which have been the object of research for many years. Silver nanoparticles have plasmonic behavior, which makes them a good candidate for new plasmonics-based photonic materials.

In most of the above-mentioned main research studies, the AgCl thin film with thickness 50 nm ≤ h ≤ 300 nm (depending on the determined TE$_n$-modes cut-off thickness), doped by Ag nanoparticles (Ag-NPs), is used as an easily available photosensitive nano-layer with waveguiding capability. The AgCl-Ag layer is deposited in vacuum, p = 10$^{-5}$ Torr, on a precisely cleaned glass substrate.
II. EXPERIMENTAL

A. Sample preparation

The AgCl thin films ($\eta_f = 2.06$) are deposited on a precisely cleaned glass substrate (substrate index of refraction $n_{S0} = 1.52$, size: 75 x 25 x 0.8 mm, a microscope glass slide), by the PVD method ($p < 10^{-5}$ Torr) from a molybdenum evaporation boat. Then, on the thin AgCl/glass planar waveguide, a very thin granular layer of silver (thickness $\sim$ 15 nm) is deposited with the same vacuum system from a tungsten evaporation boat, which provides the Ag-NPs. In this way, the Ag/AgCl/glass system acts as a planar photosensitive thin waveguide film. Other photosensitive waveguide dielectric thin films like AgBr-Ag could also be used. However, we have learned from our many experiments that the best results can be reached with AgCl nano-layers, doped by Ag-NPs.

The thickness of the AgCl layer is determined close to the cutoff thickness for TE$_0$ ($h_{0} = 50$ nm) or TE$_1$ ($h_{1} = 280$ nm) modes of the waveguide, using Eq. (2). It should be noticed that, for thick samples in which the TE$_1$-mode could be excited, the TE$_0$-mode also would be excited simultaneously. As the intensity of the TE$_0$-mode is higher than that of the TE$_1$-mode, its associated grating forms earlier, because of the PFL mechanism. The dispersion equation for asymmetric planar waveguide [8] is used to calculate the proper cutoff thicknesses for TE$_0$- and TE$_1$-modes excitation,

$$h_{m} = \frac{\lambda_{i}}{2\pi(n_{f}^{2} - n_{s}^{2})^{1/2}} \left\{ \arctg \left( \frac{(n_{f}^{2} - n_{s}^{2})^{1/2}}{m} \right) + m\pi \right\}, \tag{2}$$

where $\eta_f = 2.06$ is the index of refraction for the AgCl film, $n_{0} = 1$ is for the air, and $m$ is an integer index representing the order of the TE modes. From the calculation, we obtained $h_0 = 277.7$ nm, but for the experiment, we chose $h_0 = 280$ nm, as it is not easy to tune the vacuum deposition machine for a tenth of a nanometer.

Similar to the previous article, the Ag-NPs have an oblate spheroid shape (for example, $15 \times 30 \times 30$ nm) and uniformly distributed on the surface of the sample (Fig. 2) before the irradiation by the laser beam. The size range of thickness of the granular silver layer is about 10 nm–15 nm, and the size range of diameters of the particles is about 18 nm–60 nm. The sizes are measured using AFM images (Fig. 2).
It should be mentioned that the samples before the interaction with the laser beam are photosensitive. Thus, one should keep them in a dry and dark container before use. After the interaction with the laser beam and the formation of SPNs, in order to terminate the photosensitivity, one should remove the AgCl layer using photographic fixture solution and stabilize the SPN (which is made of Ag-NPs) by coating the formed SPN with a very thin layer of Al (thickness about 4–5 nm). For the diffraction pattern study, the thickness of the Al layer should be about 100 nm to obtain a good quality for the diffraction patterns.

B. The role of PFL in the generation of SPNs

A schematic sketch in Fig. 1(a) demonstrates the formation process of SPNs. As it was mentioned before, illumination of the sample (i.e., Ag/AgCl/Glass) by a linear polarized light (in our case, a He-Ne laser with maximum power $P_L \sim 5 \text{ mW}$ and wavelength $\lambda_i = 632.8 \text{ nm}$) leads to the excitation of $\text{TE}_n$-modes of the waveguide AgCl layer. Interference between the incident laser beam and the excited mode takes place and results in the migration of Ag-NPs from maxima to minima of the interference pattern. Thus, the SPN forms on the basis of re-distribution of Ag-NPs according to the interference pattern. After this stage, the SPN grows by the PFL mechanism, which we described in Sec. I.I. It is shown that the PFL mechanism has temporal variation and does not obey a monotonic linear rising trend. We have learned from our experiments that very large Ag-NPs (i.e., $d_{\text{Ag}} > 50 \text{ nm}$) cause lower photosensitivity of the Ag/AgCl/Glass system. In the cases for which very small Ag-NPs (i.e., $d_{\text{Ag}} < 5 \text{ nm}$) are used, the amount of available Ag-NPs is also not enough to produce a perfect SPN.

Limitation in the amount of accessible Ag-NPs is a substantial parameter for the cases when two or more SPNs are formed and compete with each other (in our case, simultaneous $\text{TE}_0$ and $\text{TE}_1$ grating formation).

C. Small angle scattering (SAS) and diffraction patterns

Because of the granular structure of the Ag layer, the $\text{TE}_n$-modes experience scattering during their propagation in the waveguide layer, which leads to a domain structure for SPNs [inset of Fig. 3(a)].
and Ref. 2]. As a consequence of the PFL mechanism, the domain structure has a temporal evolution characteristic. The $T_{E_n}$ modes scatter from the multiple borders of the domain structure, which causes appearance of a small angle scattering (SAS) pattern on a screen in front of the sample [Figs. 3(b) and 3(c)]. The SAS pattern and its evolution are explained in detail in Refs. 2, 8, and 9. For the present work, it is enough to know that the appearance of SAS patterns means that the formation of SPNs is happening and an increase of its intensity indicate the compilation process of the SPN. As it is explained in detail in Ref. 6, the SAS pattern intensity has a temporal evolution characteristic. The $T_{E_n}$-modes dominate relative to the $T_{M_n}$-modes. Therefore, because of the limited amount of available Ag-NPs for generation of associated gratings and the PFL mechanism, the $T_{E_n}$-grating formation dominates relative to the $T_{M_n}$-grating formation. Therefore, this competition in general results in a wider and more spread SAS and diffraction patterns [Figs. 3, 4, and 5(b)]. In Fig. 4, the diffraction pattern of the sample containing only the $T_{E_0}$-grating is shown. As it can be seen, the diffraction spots have an angular distribution around the maximum of diffraction, which is due to the domain structure of the induced SPN.

For the case when only the $T_{E_0}$-mode is excited, we use $d_{E_0}$ to denote the period of the associated grating. In the case of simultaneous excitation of $T_{E_0}$ and $T_{E_1}$ modes, we use $d_E$ for the period of the $T_{E_0}$-grating and $d_1$ for the period of the $T_{E_1}$-grating. That is, the period of $T_{E_n}$-gratings is different for the two above-mentioned cases (Fig. 6 in Ref. 1). That is, $d_1 > d_{E_0}$ and $d_0 < d_{E_0}$. Hence, the difference between the above-mentioned periods for thin and thick samples could be understood.

From Fig. 4 and experimental data, the distance of the diffraction pattern to the incident beam location on the screen is $x = 118.3 \text{ mm}$ (at $\theta = 0^\circ$) and the distance of the screen to the sample is $D = 85 \text{ mm}$. From Figs. 4 and 5(a) for the angle of diffraction, $\theta_0$, we have $\tan \theta_0 = \frac{x}{D}$; thus, from the equation for a grating at $\theta = 0^\circ$ at the wavelength of the $N_2$ laser probe beam, $\lambda = 377 \text{ nm}$, for the thin sample ($T_{E_0}$-grating), we obtain $d_{E_0} \sin \theta_0 = \lambda N_2$, which gives us an experimental value for the period of $T_{E_0}$-grading: $d_{E_0} = 416 \pm 2 \text{ nm}$.

The same measurement is done for the case when both $T_{E_0}$ and $T_{E_1}$-gratings are formed. In this case, we have $\theta_0 = 14^\circ$, $D = 85 \text{ mm}$, $d_0 \sin \theta_0 = \lambda N_1$, Therefore, for the $T_{E_0}$-grating, we measured $x_0 \equiv [59.06 \text{ mm}, 69.44 \text{ mm}] \pm 0.02 \text{ mm} \rightarrow d_0 \equiv [314 \text{ nm}, 340 \text{ nm}] \pm 55 \text{ nm}$ and for the $T_{E_1}$-grating, we measured $x_1 \equiv [45.42 \text{ mm}, 50.42 \text{ mm}] \pm 0.02 \text{ mm} \rightarrow d_1 \equiv [354 \text{ nm}, 370 \text{ nm}] \pm 50 \text{ nm}$. The uncertainty of the periods is related to the off-normal angle of incidence for the probe light [Fig. 5(a)] and also overlapping of $T_{E_0}$- and $T_{E_1}$-gratings.

As it can be seen from Figs. 5(b) and 5(c), the $T_{E_0}$-grating forms earlier than the $T_{E_1}$-grating. It means that the intensity of the $T_{E_0}$-mode is higher than that of $T_{E_1}$-mode (because of the PFL mechanism). In other words, the PFL rate for $T_{E_0}$-gratings is higher that than of $T_{E_1}$-gratings. From Figs. 5(a) and 5(b), we can see that the intensity of the diffraction pattern increases with exposure time, which is an experimental confirmation of the function of the PFL mechanism and self-organization of the SPN. When the quality of the $T_{E_1}$-mode associated grating is good enough, its diffraction pattern will appear [Fig. 5(b), 3] but has overlapped with the diffraction pattern of the $T_{E_0}$-grating. Thus, we observe a widespread diffraction pattern. The samples with longer exposure time exhibit perfectly separated diffraction patterns [Fig. 5(b), 4], which implies the coexistence of $T_{E_0}$- and $T_{E_1}$-generated gratings. As mentioned before, the excitation of $T_{M_n}$-modes causes more widespread diffraction patterns for both $T_{E_0}$- and $T_{E_1}$-grating diffraction patterns. On the other hand, as we discussed, $T_{E_1}$-grating caches some silver nanoparticles from the already formed $T_{E_0}$-grating.

![FIG. 3.](image-url) (a) Setup for generation of a SPN and observation of SAS patterns in real time. The domain structure is shown on the AFM image of a SPN, (b) SAS pattern of a thin sample, and (c) SAS pattern of a thick sample.
i.e., destroys it, resulting in a wider diffraction pattern of the TE₀-grating relative to that of the TE₁-grating.

The intensity distributions of the diffraction patterns of Fig. 5(b) are shown in Fig. 5(c). As it can be seen at the beginning stages of the SPN formation, a wide weak diffraction pattern is observed [Fig. 5(c), curve 1]. However, by increasing the exposure time, the intensity distribution in the larger diffraction angle rises [Fig. 5(c), curve 2], which corresponds to TE₀-grating formation. By simultaneous growing of the TE₁-grating in competition with the TE₀-grating, a widespread intensity distribution is observed, as an overlap of both corresponding diffraction patterns of TE₀- and TE₁-SPNs [Fig. 5(c), curve 3]. Curve 4 of Fig. 5(c) shows the final stage, where both TE₀- and TE₁-SPNs are formed completely and have their best quality. These results are the reconfirmation of the PFL mechanism and existence of a competition between the two nanostructures for catching the limited amount of Ag-NPs. It seems that, in stage 2 [curve 2 of Fig. 5(c)], the growth rate of formation of the TE₀-grating dominates relative to that of the TE₁-grating. Because of that, the peak of the intensity distribution of the diffraction pattern for the TE₀-grating is located at the larger diffraction angle relative to that for the TE₁-grating.

As a conclusion for this part, both TE₀- and TE₁-gratings start their self-organized formation from the very first moment of irradiation of the thick samples (h₁ = 280 nm), but due to domination of

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**FIG. 4.** Experimental setup for observing the diffraction pattern from a SPN, formed in a thin sample, in normal incidence of probe beam configuration. The top-right inset shows the diffraction pattern from a TE₀-grating.

**FIG. 5.** (a) Experimental setup for observing the diffraction pattern from a SPN, formed in a thick sample, in off-normal incidence of probe beam configuration. (b) the diffraction patterns from a sample including the TE₀-grating and the TE₁-grating, simultaneously, with different exposure times, and (c) intensity distributions of diffraction patterns of Fig. 5(b).
the TE$_0$-grating growth rate relative to the TE$_1$-grating (as the intensity of the TE$_0$-grating is higher and because the self-organization process is based on the PFL mechanism). Therefore, the diffraction pattern of the TE$_0$-grating appears earlier than that of the TE$_1$-grating. On the other hand, competition between the two dynamic nanostructures for catching (limited in amount) Ag-NPs for completing their structure leads to the variation of the intensity distribution of their diffraction patterns versus exposure time.

D. Spectroscopic studies

One of the useful methods for investigating the temporal evolution of our samples is spectroscopy. As our nanostructures are made of Ag-NPs for which surface plasmon resonance spectrum has an iconic characteristic,17 it helps us to judge their size distribution, shape, and even their arrangement (polarization spectroscopy). Although our SPNs form on the surface of the AgCl layer, the Ag-NPs have some penetration into the AgCl film, during the generation of the SPN. Thus, in general, we could take it as a nanostructure with 3D characteristics, which leads to sensitivity to the polarization sign of the probe beam.18 Besides that, dynamic self-organization of Ag-NPs according to the interference pattern results in the recording of information about the polarization state of the incident light (writing beam) within the generated SPN in the Ag/AgCl/glass system, which also makes its polarization sensitive (for the probe beam).1,5–7,9,10,12,13

On the other hand, surface plasmon resonance of Ag-NPs embedded in a matrix (in our case, the AgCl thin film) would be affected.5,6,19 A free spherical silver nanoparticle in vacuum has a characteristic absorption peak around a wavelength of 410 nm, which is due to surface plasmon resonance.17 However, under determined circumstances, an interaction with a matrix,19 any diversity in their size and shape,5,10,11,17 and also an interaction with neighboring Ag-NPs6,12 cause blue or red shift in the plasmonic absorption peak position in the absorption spectrum of the sample, relative to an isolated and free Ag-NP.

It is well-known that5–11,13,17 any arrangement of nanoparticles as a chain-like structure results in polarization sensitivity, which makes the samples dichroic (different absorption of the sample for two orthogonal polarization states of the probe beam).

We have made a series of measurements to determine the absorption spectra of our samples for different cases (Figs. 6 and 7; Spectrometer model: AvaSpec, Avantes 2048L). Extinction (absorption + scattering) spectra for both thin and thick samples are measured [Fig. 6(a)]. It can be seen that for thin samples that have larger Ag-NPs (Fig. 2), total absorption is less than that of thick samples. It could be because of the quenching of scattering of the incident light from the nanoparticles, which has more contact together due to their large sizes.6 In Ref. 6, it is shown that in the case of electrical contact of Ag-NPs, surface plasmon resonance quenches, i.e., transmittance, T, increases, which leads to less absorption of the incident light by the sample. It is more possible for the samples with larger Ag-NPs in the same amount of area for the sample with smaller ones. In addition, the thicker layer of AgCl also could have contributed to the absorption for the thick samples relative to the thin ones.

In Figs. 6(b) and 6(c), the measured absorption spectra for two orthogonal polarized probe beams at thicknesses $h_0 = 50$ nm and $h_1 = 280$ nm are shown, respectively. As it can be seen from Fig. 6(b), for the sample with a thickness of $h_0 = 50$ nm (only TE$_0$-mode excitation) at the beginning stage, there is no remarkable linear dichroism (i.e., $\Delta D = D_\parallel - D_\perp$, where $D_\parallel$ or $D_\perp = -\ln(T)$) are the optical densities of the samples for the probe beam, when its polarization vector is (i) parallel to the polarization of the incident light ($D_\parallel$), and (ii) is perpendicular to that ($D_\perp$); $T$ is the transmission coefficient, measured by the spectrometer). However, dichroism ($\Delta D$) for the thicker sample ($h_1 = 280$ nm), in which TE$_0$- and TE$_1$-modes would be excited at the same time, is obvious and changes its sign at a wavelength around 530 nm, which is very close to the surface plasmon resonance peak of embedded Ag-NPs in the dielectric AgCl matrix ($\lambda_{SPR} \sim 520$ nm).12 On the other hand, the results of our investigations show that depending on the thickness of the waveguide AgCl thin film, an increase in exposure time of writing beam could be followed by different scenarios: for thin samples, an increase of exposure time would be accompanied by an increase in...
the amount of dichroism [Fig. 7(a)], which, in general, means better organization and arrangement of the Ag-NPs. However, for thick samples [Fig. 7(b)], an increase in exposure time leads to a decrease in the order of organization and arrangement of the Ag-NPs, which is the result of overlapping of the two TE₀- and TE₁-induced gratings. Almost in all measurements for both thicknesses, the signs of dichroism are changed around the surface plasmon resonance peak of silver nanoclusters embedded in an AgCl matrix [see Figs. 6 and 7(b)]. The reason for that is explained in detail in Ref. 6. Our observations indicate that, at short exposure times of the samples for generation of SPNs, dichroism for thick samples (i.e., h₁ = 280 nm) is higher than that of thick samples (i.e., h₀ = 50 nm) [the top-left inset in Fig. 7(b)], but at longer exposure times, dichroism for thin samples increases and would be above that of thick ones. That is, the variation of difference of dichroism for thin and thick samples has a downward trend [the top-left inset in Fig. 7(b)]. It could be related to the fact that simultaneous competitive self-organization of TE₀- and TE₁-gratings is accompanied by the formation of transversal connections of the two corresponding gratings by the migrating Ag-NPs, i.e., chiral complex nanoclusters [Fig. 1(b), 2 and 3]. These transversal connections are responsible for the appearance of optical chirality of the samples. In the thick samples, the induced optical chirality proceeds more remarkably, due to the existence of competition and overlapping of two TE₀- and TE₁-SPNs.

E. AFM images

In order to investigate the structural evolution of the SPNs for both cases, we investigated the AFM images of samples with
different illumination times (Fig. 8). On the basis of AFM (AFM model: NT-MDT TS 150) images, the roughness of the samples is measured (Fig. 9). From Fig. 8, one can see that the SPNs are formed from the very first minutes of illumination and are in good accordance with the results reported in Ref. 6; their structural evolution has a variation with exposure time, which is a result of competition between the PFL mechanism and the Gaussian distribution of the incident He-Ne laser beam [for example, compare Fig. 8(a) with Figs. 8(b) and 8(e)].

From Fig. 9, it can be seen that the initial roughness of two kinds of samples (i.e., thin and thick samples) is almost the same, but after interaction with the incident laser beam, the roughness increases and after 30–45 min, it achieves its maximum. This change in roughness is more intensive for the thick sample, in which both TE0- and TE1-gratings grow simultaneously.

F. Optical activity measurements

The formation of SPNs in AgCl-Ag thin waveguide films as a result of an interaction with a low power (P_l = 5 mW) of the incident He-Ne laser beam increases the anisotropy and roughness of the samples (Fig. 9). At the same time in thick samples, where both TE0- and TE1-gratings grow, complex chiral building blocks would be formed at intermediate stages of self-organization of SPNs (Figs. 2 and 10). Both induced structural characteristics lead to the induction of optical activities in the samples as a result of interaction.2,11 As it is shown in Refs. 5 and 12, chiroptical effects in complex chiral nanoparticles with the anisotropic matrix would be more enhanced.

In order to measure the optical rotation (θ) and to determine the contributions of anisotropy and chirality in the induced optical activity of the samples, we have followed a standard method,11,12,18,25,26 whose setup is shown in Fig. 11. In this method, a linear polarized probe beam passes through the sample with an angle ϕ = 0° (ϕ is the angle between the polarization direction of the probe beam, E_p, and the polarization direction of the incident laser beam, E_o) (Fig. 11).

Before inserting the sample to measure its optical rotation power, one first puts an analyzer in front of the detector and crosses its polarization axes perpendicular to the E_p in order to obtain the minimum signal on the detector and determine the null of the setup. After inserting the sample in the setup, as it is an optical gyrotropic photonic material, the polarization direction of

![FIG. 9. Histogram of roughness for the images shown in Fig. 10.](image)

![FIG. 10. AFM images of some chiral building blocks and plane semi-helix chains made of Ag-NPs. In the right set, some chiral features are highlighted.](image)
the probe beam rotates and becomes elliptically polarized, which leads to an increase of the signal on the detector. By rotating the analyzer in the opposite direction and achieving the minimum signal again, one can measure the amount of the induced optical rotation of samples. Using a similar method, we can also determine the ellipse of polarization of the passed probe beam, which we did not study for this case, as it does not give an additional key.

Results of the measurement of an optical rotation ($\theta$) versus an angle $\varphi$ for different exposure times ($t$) are presented in Fig. 12(a), for the thick samples. As it can be seen from Fig. 12(a), this variation has a sinusoidal characteristic, and with an increase of exposure time, the total optical rotation decreases. If we do the same measurement for the thin sample where only the TE$_0$ mode could be excited, we find that the total optical rotation for this case is less than that of the thick sample [Fig. 12(b)]. We can fit a sinusoidal function to these data (curves in Fig. 12). The intercept of this curve represents the induced chirality of the SPN, and the maximum amplitude of the fitting function represents the anisotropy of the sample. As it can be seen from Figs. 12 and 13, at initial stages of irradiation (about 5 min), both chirality and anisotropy are relatively high, and by increasing the exposure time, both of them decrease. That is, in stages when both TE$_0$ and TE$_1$ gratings are forming and there is a competition between them to catch more silver nanoclusters, the chiral building blocks are abundant. At the same time, their chiroptical effect is enhanced by the induced anisotropy. However, more irradiation leads to complete formation of both SPNs, which reduces the abundance of chiral building blocks. On the other hand, thin samples almost have no chirality, even at initial stages, i.e., intercept $\sim 0^\circ$ [Figs. 12(b) and 13] and its anisotropy is much less than that of thick samples where both TE$_0$- and TE$_1$- gratings are formed [Figs. 12(a) and 13].

III. DISCUSSION

In thick samples (where both TE$_0$- and TE$_1$-modes are excited), we observed that the complex building blocks of forming SPNs have temporal evolution. AFM studies confirm the formation of the chiral building blocks (Fig. 10), which is in good agreement with the results of spectroscopic studies (Fig. 7) and measurements of optical rotation dispersion (Fig. 12). The induced chiroptical effects have temporal dependence, as well as SPN temporal evolution, and vary with exposure time.

Following the structural evolution of SPNs, the SAS patterns (Fig. 3) and diffraction patterns [Figs. 4 and 5(a)–5(c)] show temporal changes, which confirm the functioning of the PFL mechanism, the existence of domain structure of SPNs, and competition between the two formed TE$_0$- and TE$_1$- gratings.

As it was experimentally shown (Fig. 3), the SAS pattern is wider for the thick samples. It is related to the fact that in thick samples in addition to the formation of TE$_0$- and TE$_1$- gratings, the TM$_0$-grating could also exist, which leads to spreading of the SAS...
pattern. TM₀-grating, which is perpendicular to the TE₀-ones, is important in the initial stages of the SPN formation and could increase the transversal connections between the two TE₀-gratings [Fig. 1(b)], or in other words, an abundance of chiral building blocks (Fig. 10). It leads to an increase of the optical rotation due to chirality in the first minutes of irradiation (Figs. 12 and 13). However, longer exposure is accompanied with perfection of generated SPNs, which reduces the abundance of chiral nanocomplex clusters and increases the anisotropy of the samples (Figs. 7 and 13). Because of very low intensity of the TM₀-mode, governing the PFL mechanism leads to the destruction of associated SPNs with the TM₀-mode, and we will not observe its diffraction pattern but only its influence on the widening of TE₀-grating diffraction in the initial stages (Fig. 5).

The evolution of diffraction patterns in Fig. 5 shows that at first, the TE₀-grating is formed, which has a higher intensity and a larger angle of diffraction relative to the TE₁-grating and then the TE₁-grating forms.

Solving the eigenvalue equation for propagation of a TE₀-mode in a slab asymmetric waveguide, we find two solutions for the propagation coefficient for thick samples: \( \beta_{\text{TE}_0} = 0.017 \) and \( \beta_{\text{TE}_1} = 0.015 \). On the other hand, we know that the relation of the propagation coefficient of the TE₀-mode with the period of its associated grating is

\[
\text{d}_{\text{TE}_0} = \frac{2\pi}{\beta_{\text{TE}_0}}.
\]  

Therefore, in our case, \( \text{d}_{\text{TE}_0} = 370 \text{ nm} \) and \( \text{d}_{\text{TE}_1} = 419 \text{ nm} \), which are in good accordance with the experimental data presented in Subsection II C. The same calculation for the thin samples (TE₀-mode excitation only) gives us \( \text{d}_{\text{TE}_0} = 413 \text{ nm} \), which is also in good agreement with the experimental data.

FIG. 12. Results of optical rotation measurements: (a) for the thick sample with different exposure times and (b) comparison of the optical rotation of thin and thick samples for two different exposure times.

FIG. 13. (a) Variation of maxima of optical rotation angles, due to anisotropy, versus exposure time and (b) variation of offset of angle \( \theta \), which is a measure of chirality, versus exposure time, determined using Fig. 14. Both dependences are drawn for \( \phi = 0^\circ \).

FIG. 14. Variation of the absolute value of circular dichroism (CD) with exposure time for a thick sample, at different wavelengths of the probe beam.
From the diffraction equation for a grating: 
\[ d \sin \theta = \lambda \]

where \( d \) is the grating constant, \( \theta \) is the angle of diffraction, and \( \lambda \) is the wavelength of light. We can calculate the angle of diffraction for different wavelengths using the grating equation. Then, we can plot the angle of diffraction against the wavelength to obtain the grating equation graphically.

In the case of the grating equation, we have
\[ \theta = \sin^{-1} \left( \frac{1}{d} \lambda \right) \]

This equation shows that the angle of diffraction depends on the wavelength of light and the grating constant. By varying the grating constant, we can change the angle of diffraction for different wavelengths. Therefore, we can use this equation to compare the diffraction properties of different gratings.

**IV. CONCLUSION**

As a conclusion, we have carried out a series of experiments to study the formation of self-organized periodic nanostructures (SPNs) in the photosensitive slab waveguide system (Ag/AgCl/Glass) in the case of thick samples, i.e., simultaneous excitation of TE0- and TE1-modes of the waveguide. As a result of the interference the incident light with the excited TE0- and TE1-modes, Ag-NPs migrate to the minima of the interference field and the SPN generates. Each TE0-mode builds its own SPN, which competes with the other one to catch more Ag-NPs to self-organize itself according to its interference pattern. As the TE0-mode has a higher intensity, because of positive feedback of the light mechanism, its associated grating (SPN) grows earlier and faster than that of the TE1-mode. In the first minutes of exposure time, when both SPNs already exist and are competing with themselves, some Ag-NPs which are located, temporarily, in the space between the two gratings (Fig. 1) could make chiral complex nanoclusters. These chiral building blocks would be enhanced by the induced anisotropy due to the
formation of the SPNs, but in longer exposure times, they vanish because the two SPNs would be completed as much as possible and pick all migrating Ag-NPs. Thus, at longer exposure times, the abundance of chiral building blocks, which exist temporarily in the initial stages of simultaneous formation of TE$_0$- and TE$_1$-gratings, would be decreased. It is accompanied by the decrease of circular dichroism and optical gyrotropy of the samples.

Small angle scattering patterns (SAS) (Fig. 3), diffraction patterns (Figs. 4 and 5), spectroscopic studies (Figs. 6 and 7), AFM images (Figs. 8 and 10), and optical activity measurements (Figs. 11–14 and Table I) provide us with consistent data to understand and interpret the above-mentioned scenario.

Between four methods of light-induced optical gyrotropy in AgCl-Ag thin films (Refs. 11–13 and the present work), the formation of complex SPNs by successive irradiation of samples with two linear polarized lights, whose polarization vectors make an angle $\alpha$ with each other, is the most effective and controllable one. Using the above-mentioned method, one can have control on the amount and sign of the induced optical rotation on the AgCl-Ag system. Functioning in the visible region of the spectrum is one of the advantages of the induced chiral nanostructures in our experiments. The simple fabrication method is also another advantage. On the other hand, diversity in shapes of the generated chiral building blocks reduces, on average, the induced optical activity associated with the induced chirality, which may be considered as a disadvantage of nanoparticle-based induced chiroptical effects.

As information about polarization state, wavelength, and angle of incidence of the incident beam is recorded in the generated SPNs, results of the present study could be applied in optical recording and storage of optical data and also for developing a photonic material sensitive to the sign and ellipticity of the probe beam polarization. However, our suggested system is not suitable for optical switching, as the time scale of reconstruction of SPNs is in the order of minutes.

In order to investigate a possible application of SPNs formed in Ag/AgCl/Glass photosensitive waveguide nano-layers as a candidate for photonic metamaterial medium functioning in the visible region of the spectrum, more advanced experiments need to be carried out.

REFERENCES