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Optical bistability in the formation of spontaneous gratings in photosensitive AgCl–Ag waveguide films

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An experiment was conducted to create periodic structures in light-sensitive waveguide AgCl–Ag films on glass surface under irradiation by He-Ne laser beam (λ = 632.8 nm). So-called spontaneous gratings were formed from Ag granules within AgCl film on the interference pattern created by the beam and scattered waveguide modes. These gratings developed due to positive feedback: diffraction efficiency of spontaneous grating and intensity of waveguide mode are mutually amplified for account of the beam energy. Doubling of the spontaneous grating diffraction reflex has been found, and optical bistability has been shown to occur based on spatial analogy to stimulated temporal oscillations of a harmonic oscillator.

Keywords: light-sensitive materials, waveguide film, AgCl, Ag, waveguide mode, periodic structure, optical bistability.

Проведений експеримент з формування періодичних структур у світлочутливих хвилеводних плівках AgCl–Ag на поверхні скла при опромінюванні пучком He-Ne лазера (λ = 632,8 нм). На інтерференційному полі пучка та розсіяних хвилеводних мод формувались т. зв. спонтанні гратки з гранул Ag у плівці AgCl, що розвивались за рахунок позитивного зворотного зв'язку: взаємного підсилення дифракційної ефективності спонтанної гратки та інтенсивності хвилеводної моди за рахунок енергії пучка. Виявлене подвоєння дифракційного рефлексу від спонтанних граток, та на підставі просторової аналогії з часовими коливаннями гармонічного осцилятора показано, що має місце оптична бістабільність.

Ключові слова: світлочутливі матеріали, хвилеводна плівка, AgCl, Ag, хвилеводна мода, періодична структура, оптична бістабільність.

Проведен эксперимент по формированию периодических структур в светочувствительных волноводных пленках AgCl–Ag на поверхности стекла при облучении пучком He-Ne лазера (λ = 632,8 нм). На интерференционном поле пучка и рассеянных волноводных мод формировались т. наз. спонтанные решетки из гранул Ag в пленке AgCl, развивавшиеся за счет положительной обратной связи: взаимного усиления дифракционной эффективности спонтанной решетки и интенсивности волноводной моды за счет энергии пучка. Обнаружено раздвоение дифракционного рефлекса от спонтанных решеток, и на основании пространственной аналогии с временными вынужденными колебаниями гармонического осциллятора показано, что имеет место оптическая бистабильность.

Ключевые слова: светочувствительные материалы, волноводная пленка, AgCl, Ag, волноводная мода, периодическая структура, оптическая бистабильность.

Introduction

The effects of optical bistability and multistability are of permanent interest when studying nonlinear optical systems [1]. Due to significant concentration of wave energy in a small region of space when light propagation is limited by the interior of the waveguide, nonlinear effects may have a significant impact on propagation of waveguide modes [2]. It has long been known that thin silver halide films with excess of silver are photosensitive. AgCl–Ag films, which are objects of study in this work, are among them [3]. Characteristic feature of photosensitivity of composite AgCl–Ag films is transfer of silver granules to the darker areas of the interference pattern, provided silver layer is thin (about 10 nm) and therefore granular. The pattern is created either by two acting beams or by single beam interference with scattered waveguide modes. Transfer of substance is the cause of proportionality of changes not to intensity I but to energy $H = It$ where t is duration of irradiation. Therefore, researchers' attention is mainly focused on the issue of film changes dependence on H and other exposure conditions.

In this paper we reveal existence of bistability in dependence of waveguide mode intensities in AgCl–Ag films on intensity of the irradiating laser beam, not on value of exposition H.

Experiment

Waveguide silver chloride films with addition of Ag were used in the experiments. AgCl film with thickness ≈ 80 nm and Ag film (≈ 10 nm) were deposited sequentially in vacuum system at a pressure of ~ 10^{-5} mm Hg on a glass substrate. AgCl film thickness was controlled by mass of the thermally vaporized substance (30 mg), and Ag thickness was controlled by frequency change of the quartz thickness meter ($\Delta\nu$ = 15 Hz). The thickness of the resulting AgCl–Ag sample was measured using lines of equal chromatic order and was 92±2 nm.

After deposition, the sample was irradiated by 'white' light of an incandescent lamp to provide more uniform distribution of silver within the matrix of AgCl. Such a distribution of silver (in the form of granules) is indicated by an absorption band in the spectrum of the optical density. The band is associated with irradiation-induced plasma oscillations of free electrons in nanometer-sized silver granules. There is a resonance behavior of these oscillations when the frequency of light matches eigenfrequency of plasma oscillations of free electrons in the granules surrounded by silver chloride. This resonance has a maximum at $\lambda \approx 500$ nm.

The layout of the recording of spontaneous gratings (SG) is shown in Fig.1a. The photosensitive waveguide AgCl–Ag film is irradiated with a normally incident beam of He-Ne laser. Due to the interference of the incident wave with scattered waveguide modes in the film, an interference pattern emerges, and Ag granules move from maxima to minima of the interference. Prior to irradiation, silver granules are statistically uniformly distributed on the surface and inside the AgCl film. During SG recording under the laser irradiation, silver is redistributed to form a periodic structure which is a diffraction grating. Experimental evidence of SG formation are microphotographs, see [3.4].

After completion of grating recording, the sample was washed by photographic fixer to remove AgCl in order to eliminate photosensitivity. Silver granules remain on the surface of glass substrate and form the gratings which create diffraction reflections observable in autocollimation diffraction scheme (a diffracted beam propagates oppositely to the incident one). To increase brightness of the diffraction pattern, highly reflective aluminum layer was deposited onto the post-fixed sample. The test beam of He-Ne laser was focused at the center (or elsewhere) of the spot with the gratings recorded. Diffraction reflection from SG located within the particular area was observed on the screen installed across the laser beam with an aperture letting the beam through, see Fig.1b. According to the measured value of the rotation angle φ of the sample on goniometer under autocollimation conditions, SG period d was determined by formula $d = \lambda / 2\sin\varphi$ where λ = 632.8 nm is the wavelength of He-Ne laser beam which was used both to record SG and to observe diffraction patterns.

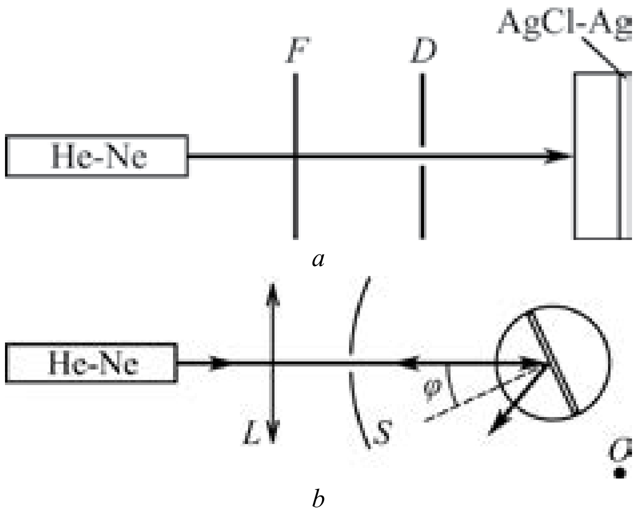


Fig.1. a) Experimental layout used to record spontaneous gratings (SG) in photosensitive waveguide AgCl–Ag films on a glass substrates using a He-Ne laser, light filter *F* and aperture *D*. b) Scheme for observation and measurements of SG periods using autocollimation diffraction after fixing film, top view. *L* is a lens to focus the beam on the sample mounted on a horizontal goniometer. *S* is a screen on which the diffraction pattern was observed in reflected light. *O* is the observation point from which the screen was photographed, see photographs in Fig.2.

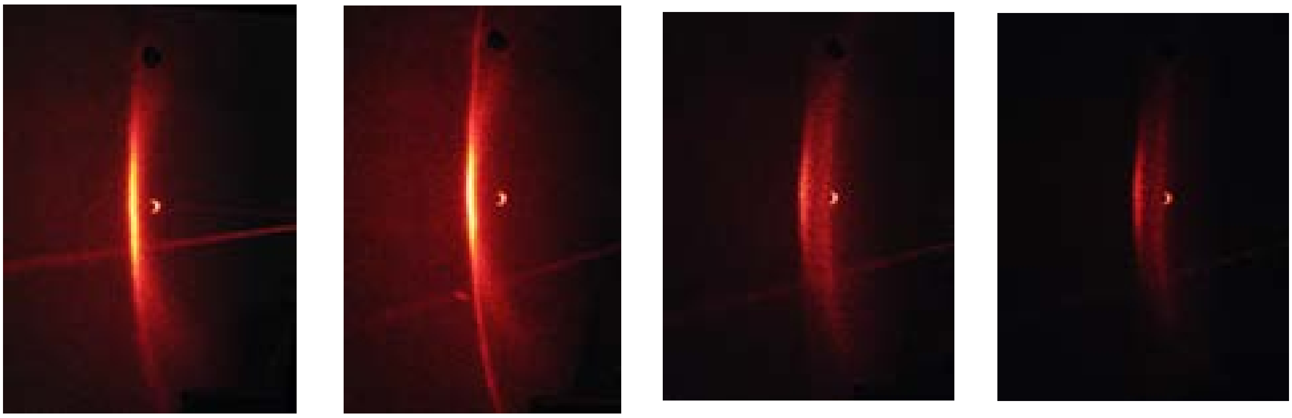
Experimental results

Both SG recording process according to the scheme in Fig.1a and observation of diffraction from SG according to the scheme Fig.1b are convenient to describe in terms of wave vectors. It is known that interference of two coherent waves with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ creates an interference pattern with vector $\mathbf{K} = \mathbf{k}_1 - \mathbf{k}_2$ in the overlapping area. On the other hand, $|\mathbf{K}| = 2\pi/d$ where d is the spatial period of the interference pattern, and direction of the vector $\mathbf{K}$ is perpendicular to the stripes of interference.

In the case of SG there is interference of the incident wave with wave vector $\mathbf{k}$ and waveguide modes with wave vector $\boldsymbol{\beta}$. Since the AgCl–Ag waveguide is thin (less than 100 nm), only projection of the wave vector $\mathbf{k}$ to the plane of the film is important when creating the interference pattern. Assuming x axis is the crossing of the plane of incidence and the plane of the film, we get an interference pattern vector $\mathbf{K}$ as

$$\vec{K} = \pm\left(\vec{\beta} - \vec{k}_x\right) . \qquad (1)$$

The vector $\boldsymbol{\beta}$ belongs to the layer plane, its value is determined by the dispersion equation for the propagation constant of the waveguide mode in a planar waveguide [5], $k_x = (2\pi/\lambda)\sin\alpha$ where α is the angle of incidence of the laser beam on the film. In the case of normal incidence



a *b* *c* *d*

Fig.2. Photographs of diffraction reflections obtained in scheme Fig.1b. In each photograph there is an aperture in the screen and vertically elongated diffraction reflections to the left of the aperture. a) The beam intensity $I = 0.75$ W/cm^2, exposure $H = 56.6$ kJ/cm^2, the test beam is focused to the center of the irradiated spot on the sample. b) $I = 7.45$ W/cm^2, $H = 58.1$ kJ/cm^2, the center of the irradiated spot. c,d) consecutive change in diffraction pattern shown in Fig.2b when shifting the test beam from the center to periphery of the spot.

of the laser wave there is $\alpha = 0$, hence $k_x = 0$, and from (1) we obtain

$$\vec{K} = \pm\vec{\beta} \, , \tag{2}$$

i.e. the wave vector of the interference pattern is determined by a 'set' of **β** vectors of the waveguide modes. A vector **β** can be directed anywhere in the waveguide layer plane because of scattering process, therefore the ends of all the vectors **K** plotted from the point of origin in the wave vector space is a circle of radius **β**. Directions of the greatest probability of scattering of TE_0 waveguide mode are perpendicular to the polarization vector $\mathbf{E}_0$ of the incident laser wave, so the 'set' of the vectors **β** and **K** is concentrated around these directions.

Since the AgCl–Ag system is photosensitive, interference patterns are recorded as gratings with 'fan'-like vectors **K**. In this case the spatial period of the gratings corresponds to the spatial period (wavelength) of the waveguide modes because of (2).

When observing diffraction from the SG in autocollimation scheme on the screen, one can see diffraction reflections from the SG demonstrating the 'fan'-like gratings. These reflections provide information both on propagation constant of waveguide modes and spread of directions of the wave vectors **β** whose appearance caused development of the SG.

Photographs in Fig.2a,b display the diffraction reflections from the SG recorded in centers of irradiated spots at different intensities of the laser beam and approximately the same energy doses. The difference in diffraction reflection brightness indicates the difference in the amplitude of the scattered waveguide modes, and their different positions (different distances to the aperture in the screen) indicate different radii of circles, i.e. the value of propagation constant β of waveguide modes depends on intensity of the incident beam.

When shifting the focused test laser beam from the center of the spot (see Fig.2b) to the periphery of the same spot (see Fig.2c,d), we can clearly see that diffraction reflection acquires double structure. We associate such a structure with manifestation of spatial bistability affecting formation of SG.

Discussion

It is possible to describe the effect of bistability on the basis of analogy of the spatial problem of waveguide mode excitation inside a film with a grating and the problem of forced oscillations of a harmonic oscillator in time, see Fig.3.

If a pump wave s falls on a waveguide with a grating, its field is modulated with the spatial frequency of the grating $\beta_S = 2\pi/d$ where d is the grating period. The modulated field of the incident wave $s = Se^{-i\beta_S x}$ acts as a driving force for the field of the waveguide mode a in the film, resulting in developing of a waveguide mode $a = Ae^{-i\beta_S x}$ with forced spatial frequency β_S. However, the waveguide mode has its 'own' resonance frequency $\beta_0(1-k\,|\,a\,|^2)$ with attenuation factor γ. The multiplier $(1-k\,|\,a\,|^2)$ with nonlinearity coefficient k takes into account dependence of the resonant propagation constant β_0 of the waveguide mode on the value of mode amplitude $|a|$, as it follows from Fig.2. As a result, we can write down the equation for coupled modes a and s [6]:

$$\frac{\mathrm{d}a}{\mathrm{d}x} = \left[-i\beta_0\left(1-k|a|^2\right)-\gamma\right]a + qs \, , \tag{3}$$

where q is coupling coefficient for waves a and s. We look

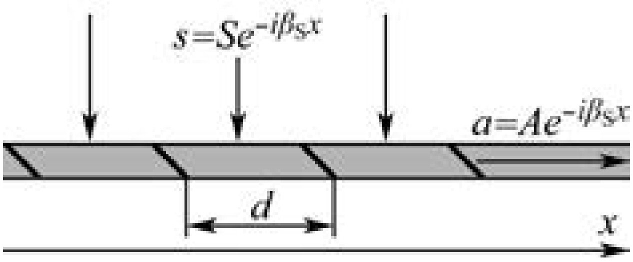


Fig.3. The scheme of spatial connection of the wave s incident on the waveguide (gray bar) with waveguide mode a through the grating with spatial frequency $\beta_S = 2\pi/d$. SG strokes are black oblique lines in the waveguide.

for a 'stationary' solution in the form $s = Se^{-i\beta_S x}$, $a = Ae^{-i\beta_S x}$, therefore

$$\begin{aligned} -i\beta_S Ae^{-i\beta_S x} &= \left[-i\beta_0\left(1-k|a|^2\right)-\gamma\right]Ae^{-i\beta_S x} + \\ +qSe^{-i\beta_S x}&\left\{-i\left[\beta_S-\beta_0\left(1-k|a|^2\right)\right]+\gamma\right\}A = qS \end{aligned}. \quad (4)$$

Similar equation must be true for the complex conjugate form, i.e.

$$\left\{i\left[\beta_S-\beta_0\left(1-k|a|^2\right)\right]+\gamma\right\}A^* = q^*S^*. \quad (5)$$

By multiplying (4) and (5), we obtain

$$\left\{\left[\beta_S-\beta_0\left(1-k|a|^2\right)\right]^2+\gamma^2\right\}|A|^2 = |qS|^2 \quad (6)$$

We introduce new variables: $I_{wm} = |A|^2 \geq 0$ is field intensity of the waveguide mode; $I_{pw} = |qS|^2 \geq 0$ is intensity of the external pump wave, at the expense of which the intensity of the waveguide mode increases, see (3). Thus (6) can be rewritten as

$$I_{pw}\left(I_{wm}\right) = I_{wm}\begin{bmatrix} k^2\beta_0^2 I_{wm}^2 + \\ +2k\beta_0\left(\beta_S-\beta_0\right)I_{wm} + \\ +\left(\beta_S-\beta_0\right)^2+\gamma^2 \end{bmatrix}. \quad (7)$$

This is a cubic equation for I_{wm}, and the shape of this curve is qualitatively shown in Fig.4 as an inverse dependence $I_{wm}(I_{pw})$.

Provided the external wave intensity is low (hence the nonlinearity may be neglected), low-intensity waveguide mode *a* is excited in the waveguide film due to the external wave *s* diffraction on the grating:

$$I_{wm} = \frac{I_{pw}}{\left(\beta_S-\beta_0\right)^2+\gamma^2}. \quad (8)$$

Dependence $I_{pw}(I_{wm})$ may be studied concerning extremums. Solving the quadratic equation in I_{wm}, obtained by equating $I'_{pw}\left(I_{wm}\right)$ to zero yields values

$$I_{wm1,2} = \frac{\left(\beta_0-\beta_S\right)^2}{3k\beta_0}\left(2 \pm \sqrt{1-\frac{3\gamma^2}{\left(\beta_0-\beta_S\right)^2}}\right). \quad (9)$$

I_{wm1} value provides the largest intensity of waveguide mode on the lower branch in Fig.4, and I_{wm2} provides the smallest one on the top branch.

At values $I_{pw} \in [I_{pw2}, I_{pw1}]$ (see Fig.4) waveguide modes with two intensity levels can be excited, which corresponds to bistability. Intermediate branch is unstable, and the states corresponding to it can not be realized [6].

Assuming absorption to be low, which means $\gamma \to 0$, we get

$$I_{wm1} \approx \frac{\beta_0-\beta_S}{3k\beta_0},\ I_{wm2} \approx \frac{\beta_0-\beta_S}{k\beta_0}. \quad (10)$$

Thus, the excitation levels of waveguide modes should differ about 3 times or more, as is observed in the images of double diffraction reflections in Fig.2c,d.

Since it is assumed that the resonant value of the propagation constant is determined as $\beta_0(1-kI_{wm})$, as it follows from (10), we obtain two values for the resonant propagation constants of the waveguide modes. The propagation constant $\beta_2 = \beta_S$ corresponds to I_{wm2} intensity level, i.e. the upper branch in Fig.4, and $\beta_1 = (2\beta_0+\beta_S)/3$ corresponds to the I_{wm1} intensity level, i.e. the lower branch. The value of β_0 is the resonant value for the 'entry' level of excitation (see (8), i.e. for the emerging SG. Grating period values ${}_0$ $2\pi/\beta_0$, $d_1 = 2\pi/\beta_1$, and $d_2 = 2\pi/\beta_2$ were calculated from angles of autocollimation diffraction and are $d_0 = 385.8$ nm, $d_1 = 392.5$ nm, and $d_2 = 403.7$ nm. On the other hand, using two of three experimentally measured periods, one can calculate the third period. So, from d_0 and d_2 the theoretical

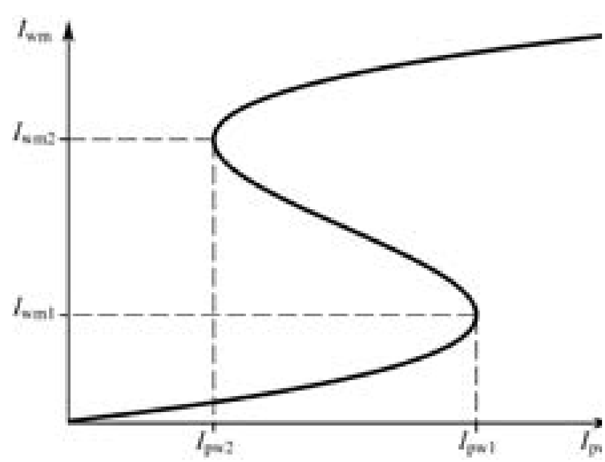


Fig.4. The qualitative form of the intensity of the waveguide mode I_{wm} on the intensity of the pump wave (laser beam) I_{pw} obtained from the equation (7).

value d_1 is

$$d_1^{\text{theor}} = \frac{6\pi}{2\beta_0 + \beta_2} = \frac{3d_0 d_2}{d_0 + 2d_2} = 391.6\,\text{nm}.$$

Divergence between d_1^{theor} and d_1 is less than 1 nm. This result may be considered as confirmation of the hypothesis of the optical bistability as the main cause of doubling the diffraction reflections observed on Fig.2c,d.

Conclusions

In this paper, an experimental study of the parameters of the plane wave diffraction on a grating in a nonlinear waveguide was conducted, and a theoretical model has been proposed to describe them. The model is based on spatial analogy with the problem of establishing forced oscillations of a harmonic oscillator. It has been shown that at diffraction on a grating in a nonlinear optical waveguide the optical bistability effect may manifest itself in dependence of the intensity of the waveguide mode on the intensity of the external incident wave. The double structure of the diffraction reflections of spontaneous gratings recorded by laser in waveguide photosensitive AgCl–Ag films is considered as a manifestation of spatial bistability effect. The calculated values of periods and intensities of grating diffraction reflections are in accordance to those observed in the experiment.

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