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KINETICS OF FORMATION OF Ag CLOTS ON AgBr MICROCRYSTALS AT THE NORMAL LASER HERSHEL EFFECT

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*It is established that clots of silver have the amorphous structure at the normal laser Hershel effect after exposure of the photographic layer SP-*1 *(photosensitivity is equal to* 6 *standard units) with low time* $t_{expos} = 10^{-5}$ *sec, and at high exposure times* (t_{expos} = 0,5 *sec) they have the crystal structure. The energy of crystallization of Ag clots equals to* $W_{min} = 10^{-8}$ *J.*

Keywords: *LASER NORMAL HERSHEL EFFECT, CLOT OF SILVER,* AgBr *MICRO-CRYSTAL, LOW-SENSITIVE PHOTOLAYER, ENERGY OF CLOT CRYSTALLIZATION.*

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1. INTRODUCTION

Photochemical release of silver from AgBr microcrystals (MC) in photoemulsion under light action should start with the appearance of separate silver atoms, whose concentration gradually increases with exposure. At the same time, these atoms interact with the formation of molecules and then amorphous clots of silver. When achieving the supersaturation in silver, metal crystalline nucleuses of silver of a critical size appear [1] that agrees with the E.O. Galashin thermodynamic theory of photographic sensitivity [2]. By this theory, accumulation of free silver in AgBr MC leads to the phase transition of amorphous silver to the crystalline state. According to the theory, increase in the interaction energy between silver atoms or molecules, which form supersaturated system with electrostatically excited electrons [3], is the main factor of crystallization.

Study of the phase transition of amorphous silver to the crystalline state is of a great importance for solid state physics [4-6], information recording and read-out by the amorphization of crystalline silver and crystallization of amorphous one, scientific photography [7, 8], since crystalline latent-image centers (LIC) arise from amorphous silver particles and play the main role in the photoprocess [9].

Increase of the supersaturation in amorphous silver in coagulation centers (CC), which appear at the laser normal Hershel effect (LNHE), leads to the three-fold increase in the photosensitivity of photoemulsion SP-1 (photosensitivity is equal to 6 standard units) without additional costs and three-fold saving in silver [10]. It is proved that LNHE has coagulation nature

[1, 11]. Crystallization of melted anthracene under the action of ultraviolet light with $\lambda = 365$ nm at exposure $t = 0{,}1$ sec is also proved in practice [12]. Light can crystallize the substance [13].

Under the action of intense laser light with $\lambda = 850$ nm and power $P = 4$ W in a pulse at surface energy density of laser light $W = 312$ J/cm^2 at LNHE amorphous silver in CC is crystallized [11, 14]. Processes, which take place at LNHE with amorphous silver in CC, depend on the density of surface light energy of laser light [11].

Nucleus of amorphous silver, which grows up, with the diameter more than 1 nm will be surely crystallized under the action of laser light with $\lambda = 440$ nm and power $P = 10$ mW at exposure $t = 0{,}5$ sec [9] that confirms the thermodynamic theory.

But growth kinetics of silver clots in photoemulsions under the mutual action of actinic and non-actinic laser radiations for different wavelengths, laser powers, and exposure times has been studied insufficiently. This paper is devoted to the investigation of this question.

2. INVESTIGATION TECHNIQUE AND EXPERIMENTAL RESULTS

To study the kinetics of the transition of amorphous silver to the crystalline Ag clots under irradiation of AgBr MC in photolayer by laser light, we have taken the system of differential equations (8) from the work [15].

We will consider each particle of silver to be a ball of the radius R which grows up due to the flows of ions and polarized atoms. Many growing particles in near-surface layer of AgBr MC appear under the action of laser light. Let $2R_m$ is the mean distance between two centers. Attraction of ions and Ag atoms to the growing clot is generated by the flow, whose initial density is equal to $J_i = n_i \cdot D_i \cdot F_i \cdot (k_b \cdot T)^{-1}$ and $J_a = n_a \cdot D_a \cdot F_a \cdot (k_b \cdot T)^{-1}$, where n_i, n_a are the numbers of vagabonding ions and Ag atoms in the unit of MC volume at the temperature of $T = 300$ K; D_i, D_a are the diffusion coefficients of ions and Ag atoms ($D_i = D_a = D$) in MC; F_i is the force acting on the ion from electron trapped on the center; F_a is the force of the electron-dipole interaction between electron trapped on the center and polarized Ag atom; k_b is the Boltzmann constant. Taking into account that attraction of the electron trapped on the center to ions and polarized silver atoms is changed by the neutralized state of the center, which remains a certain time until new electron capture. Then expressions for forces F_a and F_i should be multiplied by a fraction of time during which a particle was in the charged state, i.e. $F_i = \tau_i \cdot k \cdot e^2 / ((\tau_i + \tau_e) \cdot R^2)$ and $F_a = \alpha \cdot \tau_i \cdot k \cdot e^2 / ((\tau_I + \tau_e) \cdot R^5)$, where $k = 9 \cdot 10^9$ m/F (here e is the electron charge; τ_i is the time until capture of Ag atom by the electron trapped on the center; τ_e is the time until capture of arising electron by the trap; α is the polarizability of silver atom. Drift flows on the growing particles induce the redistribution of ions inside the ball. Therefore, we have the expressions $J_i = -D_i \operatorname{grad} n_i + n_i \cdot D_i \cdot F_i \cdot (k_b \cdot T)^{-1}$, $J_a = -D_a$,

$$\frac{dR}{dt} = -\frac{J_i(t,r) + J_a(t,r)}{n_R}\bigg|_{r=R(t)},$$

Growth rate of the particles was defined by the mass balance equation, where n_R is the atomic concentration in growing particle.

Equation

$$dn_i/dt = \sigma_V - \chi_{ie} n_i n_e - 4\pi R^2 |J_i| / (4\pi R^3{}_m/3 - 4\pi R^3/3) \qquad (1)$$

determines the temporal variation of the concentration of silver ions. σ_V describes the number of arising silver ions under the action of laser radiation in the unit of AgBr MC volume during 1 s. Term $-\chi_{ie} n_i n_e$ shows that some silver ions recombine with mobile electrons during movement to the traps and form silver atoms which have not yet joined a clot. The last term describes that electrons fixed on the traps meet with a part of silver ions, form atoms which increase the volume of silver clots during 1 s. To this end, an ion flow on the growing clot is divided by the difference between maximum volume and volume of a clot at the present moment.

Equation

$$dn_a/dt = \chi_{ie} n_i n_e - 4\pi R^2 |J_a| / (4\pi R^3{}_m/3 - 4\pi R^3/3) \qquad (2)$$

defines the change in the concentration of Ag atoms in the unit of AgBr MC volume during 1 s, which were formed at recombination of ions and electrons. The first term $-\chi_{ie} \cdot n_i \cdot n_e$ shows that some silver ions recombine with mobile electrons during movement to the traps and form silver atoms which have not yet joined a clot. The second term describes that a part of silver atoms is polarized and attracted by the fixed electron field that increases the volume of silver clots during 1 s.

Equation

$$dn_e/dt = \sigma_V - \chi_{ie} n_i n_e - \chi_{he} n_h n_e - n_e/\tau_e \qquad (3)$$

defines the change in the electron concentration. σ_V describes the number of arising silver ions under the action of laser radiation in the unit of AgBr MC volume during 1 s. The term $-\chi_{ie} n_i n_e$ determines recombination of electrons with a part of silver ions; the third term $-\chi_{he} n_h n_e$ defines recombination of electrons with holes; the last term $-n_e/\tau_e$ shows capture of some electrons by traps during 1 s.

Equation

$$dn_h/dt = \sigma_V - \chi_{he} n_h n_e \qquad (4)$$

describes with the use of light in the unit of volume the change in the hole concentration. σ_V describes the number of arising holes under the action of laser radiation in the unit of AgBr MC volume during 1 s. The term $-\chi_{he} n_h n_e$ defines recombination of electrons with holes.

We have assumed that $\chi_{ie} = \chi_{he}$, where χ_{ie} describes the recombination frequency of vagabonding silver ions and electrons; χ_{he} describes the recombination frequency of vagabonding electrons and holes. Substituting the values of J_a, J_i and taking into account the transition from R to V and expressions $\gamma = 4 \cdot \pi D \cdot k \cdot e^2 (k_b/T)^{-1}$ and $\xi = \chi_{ie} \cdot \tau_e$ in equations (1)-(4), we have obtained the following system of equations

$$\begin{cases} dn_i / dt = \left(\sigma_V \left(\xi n_h + 1\right) / \left(1 + \xi\left(n_i + n_h\right)\right)\right) - \left(\gamma n_i / \left(V_m - V\right)\right), \\ dn_a / dt = \left(\sigma_V \xi n_i / \left(1 + \xi\left(n_i + n_h\right)\right)\right) - \left(\gamma n_a 4\pi\alpha / \left(3V\left(V_m - V\right)\right)\right), \\ dn_h / dt = \sigma_V \left(\xi n_i + 1\right) / \left(\xi\left(n_i + n_h\right) + 1\right), \\ dV / dt = \left(\gamma\left(n_i + 4\pi\alpha n_a / 3V\right) / n_R\right). \end{cases} \quad (5)$$

It is theoretically proved in work [1] that the lesser LIC size, the larger costs W_v of light energy which is spent on the formation of silver LIC.

By analogy with the work [16], it was necessary to make computer models (CM) of the silver clot growth in AgBr MC of photoemulsion SP-1 (photosensitivity is equal to 6 standard units) by simultaneous exposure of two lasers with existing parameters. The first laser in all CM had the following parameters (λ is the wavelength, P is the power): $\lambda = 440$ nm, $P = 10$ mW, exposure time is $t = 0{,}5$ s. The second laser in CM had alternately the parameters: 1) $\lambda = 850$ nm, $P = 4$ W in a pulse; 2) $\lambda_1 = 1060$ nm, $P = 10$ mW, 3) $\lambda_2 = 1150$ nm, P = 10 mW. Parameters 2) and 3) correspond to the gas laser with continuous light generation. Radiation conditions correspond to the LNHE with completely superimposed in time primary and secondary exposures.

It was established by the electron microscopy method that silver crystals, which consist of tens or hundreds of Ag atoms formed mainly on the AgBr MC surface, are the LIC [17]. For the model calculations, area of AgBr MC in photoemulsion SP-1 (photosensitivity is equal to 6 standard units) was equal to $S = 10^{-12}\text{m}^2$ [15].

Taking into account the parameter values, initial conditions for the solution of the system of equations (8) were the following: $n_i = 0$, $n_a = 10^{24}$ m^{-3}, $n_h = 0$, $R = 1{,}44 \cdot 10^{-10}$ m. Parameter σ_V is the number of ions arising under the action of light and number of released holes and electrons in the unit of MC volume during 1 s. We have found: 1) for lasers $\lambda = 440$ nm, $\lambda = 850$ nm, $\lambda = 1060$ nm, $\lambda = 1150$ nm, the number of quanta radiated by laser during 1 s by the formula $N = \eta P\lambda/hc$, where h is the Planck constant; c is the speed of light in vacuum; $\eta = 0{,}5$ is the efficiency; 2) area of the laser light beam of the diameter $3 \cdot 10^{-3}$ m; 3) area of the conventional microcrystal with the size 0,5 μm × 0,5 μm. For laser with $\lambda = 850$ nm through the proportion we obtained the number of incident quanta during 0,036 s; 0,072 s; 0,144 s; 0,216 s; 0,288 s; 0,504 s; 4) through the proportion we found the number of quanta incident on the conventional microcrystal; 5) the number of quanta incident on the volume of conventional microcrystal with the size 0,5 μm × × 0,5 μm × 0,1 μm. By formulas $\sigma_{Vtot} = \sigma_{V440} + \sigma_{V1060}$, $\sigma_{Vtot} = \sigma_{V440} + \sigma_{V1150}$, $\sigma_{Vtot} = \sigma_{V440} + \sigma_{V850}$, we found $\sigma_{V440+1150} = 48 \cdot 10^{28}$ m^{-3}s^{-1}, $\sigma_{V440+1060} = 11{,}74 \cdot \cdot 10^{29}$ m^{-3}s^{-1}, $\sigma_{V440+850} = 3 \cdot 10^{28}$ m^{-3}s^{-1} for time 0,036 s, $\sigma_{V440+850} = 5 \cdot 10^{28}$ m^{-3}s^{-1} for time 0,072 s, $\sigma_{V440+850} = 7 \cdot 10^{28}$ m^{-3}s^{-1} for time 0,144 s, $\sigma_{V440+850} = 10 \cdot \cdot 10^{28}$ m^{-3}s^{-1} for time 0,216 s, $\sigma_{V440+850} = 14 \cdot 10^{28}$ m^{-3}c^{-1} for time 0,288 s, and $\sigma_{V440+850} = 2{,}4 \cdot 10^{29}$ m^{-3}s^{-1} for time 0,504 s. A program in Turbo-Pascal 7.0 was made. Results of the numerical calculations of the system of differential equations (8) were obtained by the fourth-order Runge-Kutta method.

The authors of the work [18] have proposed the model of the transition of the substance from the amorphous state to the crystalline one, where vacancy concentration n is the degree of disorder. The phase transition in the crystal deformed by vacancies is described in this work and dependence of the free energy F on the unit of volume of amorphous substance or crystal versus the vacancy concentration n is given. It is also shown in this work that consideration of the pair interaction of vacancies leads to the existence of two minimums of $F(n)$ in some temperature range. The first minimum (small n) corresponds to the crystalline phase, and the second one (large n) – to the disordered liquid or amorphous phase.

Exposure of the photolayer was performed simultaneously for two lasers 1) $\lambda = 440$ nm, $P = 10$ mW during $t = 0{,}5$ s, and for $\lambda = 1060$ nm, $P = 10$ mW during $t = 18$ s in the continuous generation mode; 2) $\lambda = 440$ nm, $P = 10$ mW during $t = 0{,}5$ s, $\lambda = 1150$ nm, $P = 10$ mW during $t = 18$ s in the continuous generation mode; 3) $\lambda = 440$ nm, $P = 10$ mW during $t = 0{,}5$ s, and $\lambda = 850$ nm, $P = 4$ W in a pulse of $2{\cdot}10^{-7}$ s. Calculations were carried out by the formula $t = \tau f d/v$, where $\tau = 2{\cdot}10^{-7}$ s is the duration of laser light pulse; $f = 10$ kHz is the number of pulses during 1 s (pulse frequency); d/v is the time during which photoplate was shifted on the length of laser beam diameter; d is the diameter of laser light beam; v is the photoplate velocity which was equal to 10^{-2} m/60 s (total time of pulses 0,036 s; 0,072 s; 0,144 s; 0,216 s; 0,288 s; 0,504 s).

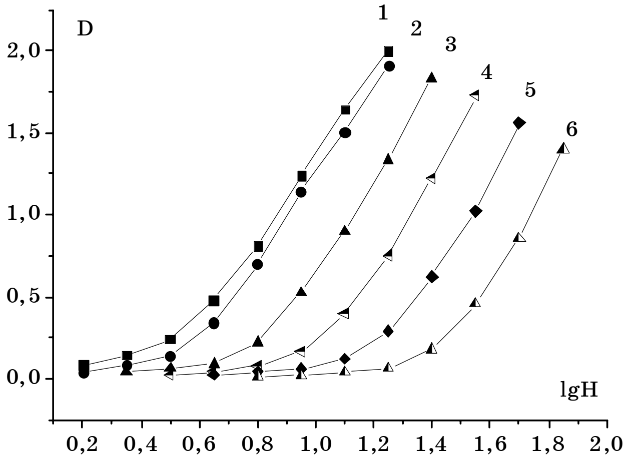

Fig. 1 *– Characteristic curves for the LNHE for photolayer SP-1 (sensitivity is equal to* 6 *units). Simultaneous exposure of photolayer was performed by laser light with* $\lambda = 440$ *nm,* $P = 10$ *mW during* $t_{440} = 0{,}5$ *s and infrared laser light with* $\lambda = 850$ *nm,* $P = 4$ *W with pulse of* $2{\cdot}10^{-7}$ *s. Curves 1 –* 0,5 *s + total exposure by pulses of* 0,036 *s; 2 –* 0,5 *s + total exposure by pulses of* 0,072 *s; 3 –* 0,5 *s + total exposure by pulses of* 0,144 *s; 4 –* 0,5 *s + total exposure by pulses of* 0,216 *s; 5 –* 0,5 *s + total exposure by pulses of* 0,288 *s; 6 –* 0,5 *s + total exposure by pulses of* 0,504 *s*

In Fig. 1 we present the characteristic curves for photolayer SP-1 (sensitivity is equal to 6 standard units) for the condition 3) with $\lambda = 440$ nm and power $P = 10$ mW, and $\lambda = 850$ nm and $P = 4$ W in a pulse of $2{\cdot}10^{-7}$ s for LNHE at different total time of pulses by laser light. With the increase in the total time of pulses characteristic curves 2-6 were shifted to the right along the lg$H$-axis with respect to the characteristic curve 1 obtained under the condition 3) for 0,036 s.

3. DISCUSSION OF THE RESULTS

For each studied light wavelength and laser power, minimum and maximum values of the silver clot volume V were shown on the computer screen. These values and numerical values of N_i and time of their formation were entered in the Table 1. These values were checked using the graphs. For example, we dropped a perpendicular to the time-axis 30 lg(t) in the point corresponding to the exposure time of 0,5 s, which was prolonged to the cross point with curve 1 (clot volume). From this cross point we plotted a horizontal line to the section with the ordinate axis and found, for example, number 220. Then by the formula $-$ $10\lg(V_{max}) = 220$, $\lg(V_{max}) = -22$ we obtained the silver clot volume $V_{max} = 10^{-22}$ m^3. Similarly we found the values of N_i and entered them in the Table 1.

Table 1 *– The normal laser Hershel effect*

Laser light parameters		Total time of laser light pulses $\sum t_{pul}$ (s)	Small exposure time by laser light 10^{-5} s			Large exposure time by laser light 0,5 s				Crystallization energy of Ag clot for $t = 0{,}5$ s,$\cdot10^{-8}$ (J)
Wavelength λ (nm)	Optical power in a pulse P (W)		Minimum Ag clot volume $V_{min}\cdot10^{-24}$ (m^3)	Clot diameter in comparison with $d_{cr} = 10\cdot10^{-10}$ m, $2R = d_{cr}$ (10^{-10} m)	Amorphous (A) or crystalline (C) silver clot		Clot diameter in comparison with $d_{cr} = 10\cdot10^{-10}$ m, $2R = d_{cr}$ (10^{-10} m)	Maximum Ag clot volume $V_{max}\cdot10^{-21}$ (m^3)	Number of silver ions N_i at exposure time $t = 0{,}5$ s, (m^{-3})	
850	4	0,036	14	$0{,}67 < d_{cr}$	A	C	$620 > d_{cr}$	10	$5{,}87\cdot10^{25}$	0,25
850	4	0,072	23	$1{,}95 < d_{cr}$	A	C	$844 > d_{cr}$	17	$4{,}8\cdot10^{27}$	0,5
850	4	0,144	32	$2{,}28 < d_{cr}$	A	C	$1064{,}6 > d_{cr}$	26	$4{,}85\cdot10^{27}$	1
850	4	0,216	47	$2{,}05 < d_{cr}$	A	C	$1149 > d_{cr}$	32	$7{,}78\cdot10^{25}$	1,5
850	4	0,288	66	$2{,}31 < d_{cr}$	A	C	$1214{,}2 > d_{cr}$	40	$1{,}31\cdot10^{25}$	2
850	4	0,504	1,1	$2{,}46 < d_{cr}$	A	C	$1340 > d_{cr}$	41,8	$8{,}8\cdot10^{24}$	3,5
1060	10^{-2}	18	5,4	$1{,}34 < d_{cr}$	A	C	$267 > d_{cr}$	4,4	$3{,}78\cdot10^{29}$	0,3
1150	10^{-2}	18	2,3	$1{,}96 < d_{cr}$	A	C	$198 > d_{cr}$	1,0	$1{,}47\cdot10^{29}$	0,3

As seen from Table 1, with the increase in the total exposure time (t_{pul}) by the infrared laser light with $\lambda = 850$ nm, Ag clot volume increases from $V_{min} = 1{,}9\cdot10^{-22}$ m^3 to $V_{max} = 4{,}1\cdot10^{-21}$ m^3. Number of atoms N_a in Ag clot increases from $1{,}8\cdot10^{28}$ m^{-3} to $4\cdot10^{28}$ m^{-3} for laser light with $\lambda = 850$ nm at exposure by infrared light during $t_{exp} = 0{,}504$ s, and for laser light lengths $\lambda_1 = 1060$ nm, $\lambda_2 = 1150$ nm – $N_{a1060} = 4{,}3\cdot10^{29}$ m^{-3}, $N_{a1150} = 2{,}0\cdot10^{25}$ m^{-3}, respectively. For $\lambda = 850$ nm the number of Ag$^+$ ions decreases from the value of $6{,}34\cdot10^{27}$ m^{-3} to $8{,}8\cdot10^{24}$ m^{-3} that agrees with the thermodynamic theory of photographic sensitivity about the fact that a part of Ag^0 atoms, formed due to the recombination of Ag$^+$ ions and electrons through the polarization of atoms by electric field of an electron, passes to Ag clot [19], and for the values $\lambda_1 = 1060$ nm and $\lambda_2 = 1150$ nm it is not change and equal to from the value of $3{,}7\cdot10^{29}$ m^{-3} to the value of $2{,}3\cdot10^{29}$ m^{-3}.

With the increase in the total time of laser light pulses ($\lambda = 850$ nm), the energy of clot crystallization increases (10^{-8} J) that agrees with the work [20]. With the increase in the energy of light quanta and the number of absorbed ones by AgBr MC, crystallization, probably, becomes easier. It is established in the works [9, 15] that silver particle with the diameter of 1 nm will be surely crystallized at its further growth. At exposure by infrared laser light during 0,5 s on the scale – 30lg(t), the number of ions N_i is separated from the curve of holes N_d, and increase in the number of Ag ions from the value of $4{,}8\cdot10^{27}$ m^{-3} to the value of $1{,}47\cdot10^{29}$ m^{-3} occurs. Slowing-down of recombination of Ag$^+$ ions and free electrons is observed, since molecules Br$_2$ are formed from the excited bromine atoms [21]. During formation each molecule attaches two electrons with opposite spins, and the number of electrons which were formed in the reaction $\mathrm{AgBr} + h\nu \rightarrow \mathrm{Ag}^+ + \mathrm{Br}^- + \mathrm{e}$ decreases. Without electrons Ag$^+$ ions can not recombine and form Ag^0 atoms. Calculation results are represented in Table 1.

Thus, in the work we have proved the validity of the theoretical conclusion [1] that the lesser LIC size, the larger costs of light energy spent on the formation of silver LIC. Numerical value of the light energy is obtained. The value of the LIC crystallization energy at LNHE is equal to 10^{-8} J.

4. CONCLUSIONS

In the present work, using the results of model calculations of the growth kinetics of Ag clots under simultaneous action of actinic and non-actinic laser radiations, we can conclude the following:

1. At LNHE for small time $t_{exp} = 10^{-5}$ s Ag clots have the amorphous structure, and for $t_{exp} = 0{,}5$ s silver clots have the crystalline structure.
2. Crystallization energy of Ag clots at LNHE is equal to 10^{-8} J.
3. Validity of the theoretical conclusion of work [1] that with the increase in the light energy costs on the formation of one silver LIC the LIC size decreases is proved. This size for LNHE is determined.
4. Results of model calculations agree with the items of the theory [1, 2] as well as with the results of the works [5, 9, 17].

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