ON THE THEORY OF FLUCTUATIONS IN THE SENSITIVITY OF CRYSTALS OF NUCLEAR PHOTOGRAPHIC EMULSIONS

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Abstract

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ON THE THEORY OF FLUCTUATIONS IN THE SENSITIVITY OF CRYSTALS OF NUCLEAR PHOTOGRAPHIC EMULSIONS

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The formation of a latent image in the crystals of a photographic emulsion is inevitably statistical in character. This applies, in particular, to the case in which the cause of the formation of the latent image is the passage of an ionizing particle through the emulsion. In this process, both the ionizing action of the particles and the properties of the emulsion crystals are subject to fluctuations. An attempt to construct a theory of ionization fluctuations in an emulsion consisting of identical crystals (or of several groups, within each of which all crystals are identical), undertaken by K. S. Bogomolov ([1,2]), proved unsuccessful, since it led to results contradicting experiment ([3]). Moreover, the failures of this theory show that ionization fluctuations, even for relativistic particles, generally play a small role, and that the experimentally observed fluctuations of the mean density of particle tracks in an emulsion should rather be attributed to fluctuations in the sensitivity of individual crystals. The corresponding version of the theory is set forth below.

We shall express the sensitivity of crystals by the number n of Ag atoms that must be formed in a crystal in order to make it developable. Any sensitization of an emulsion with respect to certain ionizing particles decreases n, and fluctuations of sensitivity are at the same time fluctuations of n. Suppose that before sensitization of the emulsion the developed track of a particle had a density $\nu_{\text{n}}(\geq 0)$ grains per unit length of track, and after sensitization $\nu_{\text{c}}(> \nu_{\text{n}})$. From this one can find ([4,5]) the decrease (Δn) in the number n of electrons formed by the particle in an individual crystal and, in total, in all crystals affected by it over some segment of the track; the number ν_0 of crystals lying per unit path of the particle, needed for the calculation, is given by Zhdanov' s formula. Thus, in order to impart developability to the crystals after sensitization, it was necessary to form, per unit path, $\nu_0 \Delta n$ fewer electrons. If electron losses are neglected, i.e., if it is assumed that each electron formed ultimately led to the formation of an Ag atom, then one may say that it was necessary to form $\nu_0 \Delta n$ fewer Ag atoms.

Thus, the action of the sensitizer may be regarded as the equivalent formation of $\nu_0 \Delta n$ Ag atoms without exposure (in reductive sensitization, for example, these

atoms are actually formed). Excluding from consideration the fraction ν_{n}/ν_0 of crystals which would have become developable even before sensitization, we must now consider the distribution of these $\nu_0(1-\nu_{\text{n}}/\nu_0)\Delta n$ Ag atoms over the $\nu_0-\nu_{\text{n}}$ crystals. If they were distributed uniformly, either all $\nu_0-\nu_{\text{n}}$ crystals or none of them would become developable; owing to fluctuations, however, only those $\nu_{\text{c}}-\nu_{\text{n}}$ crystals in which a sufficient number of Ag atoms was formed acquired developability. In order to determine by comparison with experiment what number of Ag atoms (regarded as concentrated at one center) is sufficient for developability, it is necessary to have the solution of the following problem: for an arbitrary distribution of r ···

different balls (Ag atoms) among s boxes (crystals): what, on average, is the fraction of the total number of boxes constituted by boxes containing not less than a certain number of balls?

Let us denote the fraction of boxes containing not less than i balls by α_i. Then the values α_i, as can be shown, are given by the formula

$$\alpha_i = \frac{\binom{s+r-i-1}{r-i}}{\binom{s+r-1}{r}}, \tag{1a}$$

where the symbol $\binom{a}{b}$ denotes the binomial coefficient C_a^b. Since in the data used in our problem r and s are referred to the unit of length of the track whose choice has been made, the quantities α_i should be replaced by their asymptotic values for $r, s \to \infty$, $r/s = \lambda = \text{const}$. It is easy to see that then

$$\alpha_i = \left(\frac{\lambda}{1+\lambda}\right)^i. \tag{1b}$$

From formulas (1) one computes the fraction of boxes containing exactly i balls:

$$\Delta\alpha_i = \alpha_i - \alpha_{i+1} = \frac{\binom{s+r-i-2}{r-i}}{\binom{s+r-1}{r}}, \tag{2a}$$

and also its asymptotic value

$$\Delta\alpha_i = \frac{\lambda^i}{(1+\lambda)^{i+1}} = \frac{\alpha_{i+1}}{\lambda}. \tag{2b}$$

Formulas (2) express the distribution of crystals by sensitivity; moreover, owing to the indistinguishability of the balls, it is geometric (see, for example, ([6])), and not Poissonian.

If each crystal has N centers of sensitivity, at each of which groups of Ag atoms can be formed, then in distributing atoms among the individual centers it is

necessary to take into account that the presence of a group of Ag atoms at one of the centers is useless if another center in the same crystal already contains such a group. Therefore it is necessary to consider the distribution of r balls among s groups of boxes (of N each), in which at least one of the boxes of each group contains not less than i balls. The fraction of groups of boxes satisfying the stated condition is given by the following formula (we give only the asymptotic value), which passes into (1b) for $N = 1$:

$$\alpha_i(N) = \left(\frac{\lambda}{1+\lambda}\right)^{N(i-1)+1} + \frac{1}{\lambda}\sum_{l=i}^{N(i-1)}\left(\frac{\lambda}{1+\lambda}\right)^{l+1} W_{li}, \tag{3}$$

where

$$W_{li} = \sum_{k=1}^{N}(-1)^{k-1}\frac{\binom{N}{k}\binom{N+l-ik-1}{l-ik}}{\binom{N+l-1}{l}}.$$

Here the first term takes into account those crystals in which the number of atoms is sufficiently large that at least one center must necessarily have not less than i Ag atoms, while the second term takes into account those crystals where a group of i or more atoms at one center is possible but not necessary, W_{li} being the probability of formation of such a group.

Before presenting the results of comparison of the formulas obtained with experimental data, let us note that these formulas contain not a single arbitrary parameter. Therefore the degree of agreement with experiment that will now be demonstrated is in itself more important for the fundamental confirmation of our model than an improvement of this agreement by introducing various semiempirical coefficients.

Formulas (1) can be applied to the especially fine-grained emulsion of Perfilov ([7]). The crystals of this emulsion certainly have only one ···

toward the sensitivity center each (if they have one at all), and the electron losses in them are negligibly small, since the crystal diameter ($\sim 0.08\,\mu$) does not exceed the free path length of electrons in crystals. In the course of ripening this emulsion is brought to moderate relativistic sensitivity (track density $\nu_{\text{H}} \simeq 25$–30 grains per $100\,\mu$ of track), and then is additionally sensitized with triethanolamine, whereby a track density ν_{c} of up to 60 grains per $100\,\mu$ is attained. The second sensitization, as is easy to calculate (taking $\nu_0 = 870$ grains per $100\,\mu$ (7)), leads to a decrease in the number of electrons (and therefore also of Ag atoms, since electron losses are negligible) by only $\Delta n = 0.16$. This result, in the absence of fluctuations, would be physically meaningless; however, for a random distribution $\nu_0(1-\nu_{\text{H}}/\nu_0)\Delta n \simeq 140$ Ag atoms over $\nu_0 - \nu_{\text{H}} \simeq 840$ crystals ($\lambda = 1/6$), there are $\alpha_1 = 14\%$ of crystals that have received at least one Ag atom, and $\alpha_2 = 2\%$ of crystals that have received no fewer than two

atoms. The latter is rather close to the increase in track density in the experiment ($\sim 3\%$ of ν_0) and shows that the result of the action of triethanolamine is a group of at least two atoms, i.e. a subcenter, as also follows from other data (3,8).

Formulas (1) are also applicable to the increase in fog upon secondary sensitization of the same emulsion. It is about 2–4 grains per $1000\,\mu^3$, i.e. about 10^{-6} of the total number of crystals. For $\lambda = 1/6$, such a fraction is made up by crystals for which $i = 7$, i.e. those that have received 7 or more Ag atoms; since they were developed without exposure, the minimum size of the development center for this emulsion is 7 atoms, in good agreement with data for other relativistic emulsions (9).

For the coarser-grained P-NIKFI emulsion ($\bar{d} = 0.28\,\mu$) formula (3) should be applied, since it has been established by the electron-microscopic method (10) that its crystals usually have 2–3 sensitivity centers. Additional sensitization with triethanolamine increases the density of relativistic tracks in this emulsion from 30–35 to 60–80, and in individual cases to 100 grains per $100\,\mu$ (the latter is attainable only with a simultaneous considerable increase in fog) (11). Taking ν_0 equal to ~ 300 grains per $100\,\mu$ (1,2), $\nu_H = 30$, $\nu_c = 60$–80, we have $r = 500$–800, $s = 270$, $\lambda = 2$–3. Then an increase in density of ~ 10–20% of ν_0, observed in the experiment, corresponds to $i \simeq 4$–5, which is at least twice as large as i for Perfilyev' s emulsion. This, however, becomes readily understandable if one takes into account that in larger crystals electron losses are large, amounting to no less than 50%. Then the number of Ag atoms at the center is no longer i, but is only ~ 2, i.e. subcenters are formed here as well. The increase in fog is 1–2 grains per $1000\,\mu^3$ (about 10^{-4} of the total number of crystals) at $\nu_c = 60$–80, $\lambda = 2$–3, and rapidly increases to 5–6 and more grains per $1000\,\mu^3$ ($\sim 10^{-3}$ of the total number of crystals) with a further increase in track density, for example to $\nu_c = 100$ (in this case $r = 1100$, $\lambda = 4$). These data agree with formula (3) for $i \simeq 8$–12, which also does not contradict the available experimental data (9).

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Note: Figure translations are in progress. See original paper for figures.