

LXXXV. *Quantum Theory of Photographic Exposure.*  
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IN the present paper an account will be given of some additional experimental tests of the light-quantum theory of photographic exposure proposed in the first paper of the same title†, and some further theoretical formulæ will be deduced from the fundamental one given in that paper. First of all, however, due mention must be made of certain very valuable experimental investigations, since published by Svedberg, which seem again to corroborate the theory, also of a paper by Svedberg and Andersson published somewhat earlier, but not brought to our notice until the first paper had been dispatched for publication.

1. Concerning "The Effect of Light," Svedberg and Andersson's paper (Phot. Journal, August 1921, p. 325), dealing under that head with only a very few size-classes of grains (each class, moreover, of a very considerable breadth), contains only the qualitative though definite conclusion that 'for equal exposure the percentage of developable grains is always greater in the class of larger grains.' The quantitative, viz. exponential dependence of this percentage upon the size (area) of the grains, is obtained and well verified experimentally in the case of bombardment by  $\alpha$ -rays, Kinoshita's experiments of 1910 having made it very probable that each silver halide grain hit by an  $\alpha$ -particle is made developable. The latter being granted and the discrete nature of  $\alpha$ -rays being a palpably established fact, the validity of the exponential formula, in our symbols  $k = N(1 - e^{-na})$ , had to follow as a necessary consequence. Its verification is properly a verification of Kinoshita's statement, and by having thus tested it experimentally Svedberg and Andersson have certainly done an important piece of work, especially as Kinoshita's result was contested by St. Meyer and v. Schweidler. In the next section analogous experiments with  $\beta$ -rays are described, but the results thus far obtained are not conclusive apart from enabling the authors to state that one or two  $\beta$ -particles striking a grain do not as a rule make it developable. Finally, returning once more to the effect of light (p. 332),

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† L. Silberstein, Phil. Mag. July 1922, p. 257.

the authors remark only briefly that an analogous conception might also assist in the interpretation of the mode of its action ; but add that if the quantum hypothesis be assumed, "the difficulty arises that the real blackening curve has not the exponential form prescribed by this hypothesis if we suppose each halide grain to be made developable when struck by a single light quantum." They seem to forget that the simple exponential formula yielded by a quantum theory relates to the case of equal grains, which is not that of real emulsions, and that in order to obtain the blackening curve (say density  $D$  plotted against the logarithm of exposure) that elementary formula has to be integrated over the range of sizes, which presupposes the knowledge of the frequency curve of the emulsion, and the somewhat intricate question of the relation between the photographic "density" and the total of blackened areas has to be treated\*. The latter question, simple though it be for one-layered coatings, becomes particularly intricate in the usual case of many layers of grains. It is for this reason that the best way of testing a similar theory consists in microphotographic counts and planimetric measurements of the individual grains. At any rate, Svedberg and Andersson propose to turn to another more complicated assumption† which, they expect, "will actually predict a blackening curve of S-shape." They propose to discuss this possibility on another occasion.

The second of the papers alluded to, due to Professor Svedberg himself (Phot. Journal, April 1922, p. 186), has a more direct bearing upon our subject, and may turn out to supplement our own tests by offering, as it were, an intermediate link in the conjectured mechanism of the action of impinging quanta or light darts. In this paper Svedberg proposes to explain the behaviour of the grains noted in his preceding paper by a single hypothesis, and to test the latter directly. His hypothesis is that the product of the light action on the halide grain consists of "small *centres* distributed through the grain or through the light-affected part of the grain according to the laws of chance," and that a grain will become developed if it contains one or more such centres. If  $\nu$  be the average number of centres per grain, the percentage probability that a grain will contain at least one centre (and will therefore be developable) is  $P = 100 (1 - e^{-\nu})$ .

\* Concrete examples of such a kind will be treated in the third paper on our subject.

† Namely, that a certain minimum number of quanta must strike the grain within a certain maximum part of its area in order to "build up a silver nucleus large enough to act as a reduction centre."

Now, it would be enough to assume that these centres are produced by discrete light-quanta impinging upon the grain, and the formula  $P=100(1-e^{-na})$  would follow at once. (For, if  $n$  be the number of light-quanta per unit area, and  $a$  the area of a grain,  $v=na$ .) But Svedberg does not make this assumption\*, and devotes instead the remainder of his paper to testing directly the above formula for the occurrence of at least one centre and the corresponding chance formula for the percentage number of grains having no centres, of those having one or two or three centres, etc., having succeeded in making these centres or, in Svedberg's own words, "the nuclei corresponding to the developable centres," visible and accessible to measurement. For details of these elegant experiments the reader must be referred to the original paper. Here it will be enough to say that the recorded "dots" or visible traces of those centres were found distributed very much in accordance with the probability formulæ, namely, in one experiment with light and one with X-rays. Only two *size*-classes of grains were treated in each of these experiments, and with regard to the dependence upon *exposure* Professor Svedberg (p. 192) has thus far only roughly stated that the percentage number of developable grains "increases approximately exponentially as function of exposure," at least for normal and for over-exposures in the case of light (and probably for all exposures in the case of X-rays) though not for under-exposure to light. The paper is concluded by the remark that to account for the deviation from the exponential formula in the case of low light-exposure, we should probably have to adopt the quantum point of view, and that in the case of light (a quantum of visible light containing 5000 times less energy than an X-ray quantum) "several quanta would have to be absorbed very near one another to form a developable centre." Such a view, however, can easily be shown to be untenable. At any rate, Professor Svedberg proposes to test it by experimental investigations which are planned in this direction.

\* In the discussion which followed upon the reading of Svedberg's paper, Prof. T. M. Lowry mentioned such an assumption of a "bombardment by light corpuscles" as the simplest interpretation of Svedberg's photographs (of the "centres"). Other speakers, however, were rather hostile to such a view, and Mr. B. V. Storr considered it even equally conceivable that the "centres" distributed haphazardly might be present before the light action, but such a state of things would have hardly escaped Svedberg's notice. At any rate, Professor Svedberg will no doubt meet Mr. Storr's objection by appropriate control experiments. Control experiments of such a kind, viz. desensitizing experiments, are now being made by Sheppard and Wightman.

The existence of the aforesaid "centres" as seats of incipient development, around which the developer's action gradually spreads, has been known for some years\*, and has been observed, among others, by Trivelli. But the important discovery that these centres are haphazardly distributed is entirely due to Professor Svedberg. If his results are ultimately confirmed by further experiments, especially for a series of different exposures, it will be possible to consider these centres as an intermediate link in the theory proposed in our first paper (the centres marking the spots where the grains were hit by the light-darts). In the meantime, however, our further tests have to be conducted by considering the *last link* of the chain, *i. e.*, by counting the grains of each size-class affected and ultimately developed.

2. Before passing on to the description of our further experimental results, a few words must be said in defence of the property attributed in our first paper to *clumps* (aggregates) of grains which apart from some single grains constituted our chief material. An explanation seems the more necessary, as another recent paper by Svedberg† contains results apparently clashing with what we believe to be the behaviour of clumps with respect to light. The property assumed by us, as the expression of experimental facts, was that a *clump*, *i. e.*, an aggregate of grains in contact with one another, behaves as a photographic unit, by which is meant that if any one of its component grains is made developable, *the whole clump* will be reduced by a sufficiently long development. We have since been able to test this behaviour in a variety of ways.

On the other hand, Svedberg concludes from his experiments that there is no transference of reducibility (developability) from one grain to another "in direct contact" with it. (See especially p. 184, *loc. cit.*)

This apparent discrepancy seem to be due to the circumstance that Professor Svedberg worked with an emulsion (a single kind only) consisting of rather small and almost spherical grains, whereas our material, and especially the so-called W-12-C experimental emulsion, with which all the work in question is being done, consists predominantly of large and very thin, flat polygonal plates or tablets which are in mutual contact either along a whole edge or, still more intimately, are partly piled upon or overlapping each other.

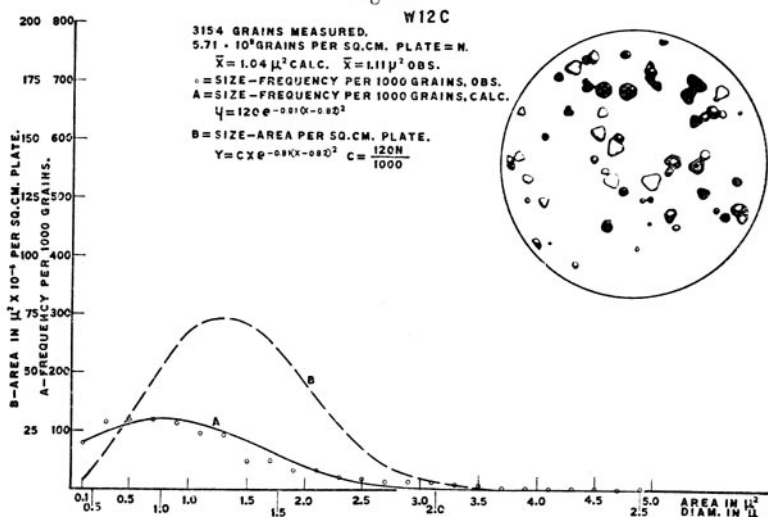
\* Cf. M. B. Hodgson, Journ. Franklin Inst., November 1917.

† On "The Reducibility of the Individual Halide Grains," Phot. Journal, 1922, pp. 183-186.

The fine spherical grains of Svedberg could have only at the utmost a point contact, and this might not have been intimate enough. It is even credible that in view of the Brownian motion of these minute bodies there was actually no permanent contact between them, as becomes very likely from Svedberg's remark on page 185, that "even over such a small distance as 1 micron no noticeable transport of silver ions takes place."

At any rate, we have found in our case the property of clumps as units well verified. Without attempting to reproduce in this place all our available evidence\*, we may support and illustrate the said principle by the following data. Fig. 1 represents the frequency curve and, in the

Fig. 1.



inset, a microgram of a sample of grains of the aforesaid W-12-C emulsion. This emulsion was spread over the glass plate in a single layer so as to obtain the maximum number of grains per unit area with the least possible overlapping. The emulsion, after the coating, was kept in its liquid state long enough to enable the majority of the grains to settle with their flat faces on the surface of the glass. Under these circumstances they, and especially the larger grains, form numerous clumps of from 2 up to 33 grains, as will be

\* Discussed in a paper just sent to Phot. Journ. by Trivelli, Righter, and Sheppard. [This paper has since been published in Phot. Journal for September 1922, p. 407.]

manifest from Fig. 2, curve marked *N*. After exposure and development the clumping of the survived grains was determined all over again and is represented by the curve marked *N-K*; the curve marked *K* is the difference of these two curves and represents the clumps affected by light.

Fig. 2.

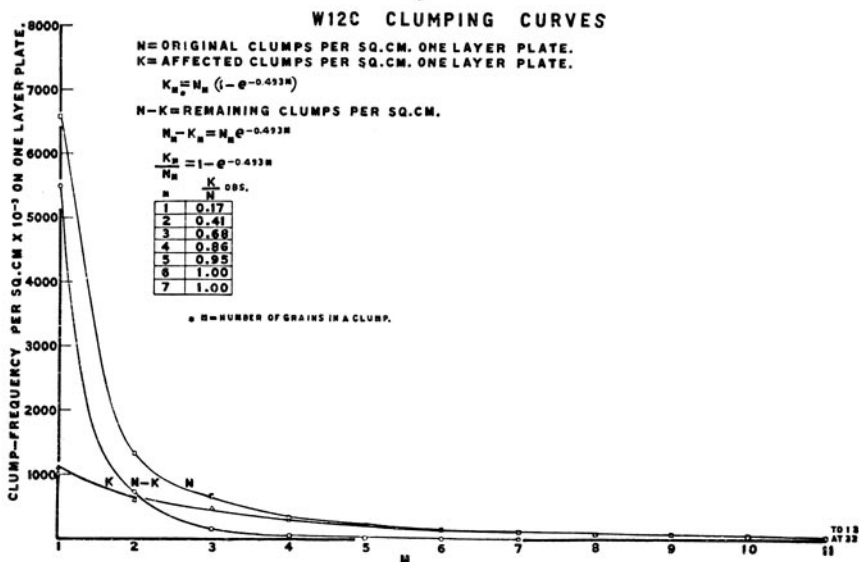
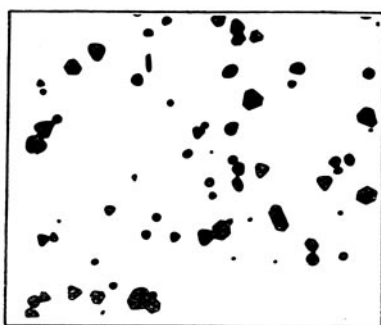
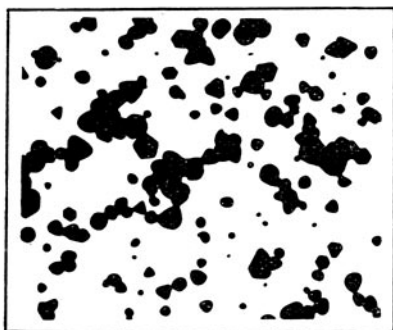


Fig. 3 a.

Fig. 3 b.



This would suffice perhaps by itself to show that our clumps behave as photographic units. But additional evidence is afforded by figs. 3a and 3b, in which all the individuals were carefully blackened by hand on a microgram originally enlarged 10,000 times; the former of these figures refers to the original unexposed one-layer grains, and the latter to

the grains surviving after exposure and development. A glance will show that the majority of clumps, and especially the larger ones, are removed entirely. Of such pairs of samples as figs. 3*a* and 3*b*, about forty were made, and the behaviour was always typically the same. Further and more direct experimental tests of the adopted clump principle are now in progress, notwithstanding that we have but little doubt about its correctness, always, of course, in relation to the material which we are using. And we feel sure that the same principle can be firmly relied upon in what follows.

3. Let us recall that the theoretical values of the percentage number  $y=100k/N$  of clumps affected, as given in the fourth column of the table in our first paper, were calculated by means of formula (12),

$$\log \left( 1 - \frac{k}{N} \right) = -na[1 - \sqrt{\sigma/a}]^2, \dots \quad (12)$$

with the values of the parameters

$$\left. \begin{aligned} n &= 0.572 \text{ per } \mu^2 \\ \sigma &= 0.097 \mu^2, \end{aligned} \right\} \dots \quad (12a)$$

the meaning of all the symbols being as before. The agreement of these values with the observed ones, ranging over 33 classes of grains and clumps, was excellent, thus proving, at any rate, the essential correctness of the formula as far as the dependence on size (area)  $a$  goes.

To test it with regard to the exposure or  $n$ , portions of the same plate were subjected to the action of the same light source, *cæteris paribus*, for one-half, and for one-quarter of the time of the original exposure. The same method of evaluating  $N$  and  $k$  being adopted as before, the results tabulated below under  $y_{\text{obs.}}$  were obtained. Now, without even taking the trouble of retouching the values of the parameters in adaptation to the new observations,  $\sigma$  was taken as in (12*a*) and  $n$  equal to one-half and to one-quarter of its original value, respectively. Since the exposure is, at any rate, proportional to  $n$ , our formula with these  $n$ -values should represent the two new sets of observations. The following table gives in the first row the number of grains in a clump\*, and in the second row the average area  $a$  of each class of clumps, in square microns, as before; the third and

\* Starting from 2, since with these weaker exposures reliable counts of single grains affected could not be secured.

the fifth rows contain the percentage numbers of clumps affected calculated by (12) with  $\sigma=0.097$  and

$$(I.) \quad n = \frac{0.572}{2} = 0.286 \text{ per } \mu^2$$

and  $(II.) \quad n = \frac{0.572}{4} = 0.143,$

respectively, and the fourth and the last rows those observed. The last but one column refers to clumps of 12 and 32 grains, and the fact that almost all of these have been affected ( $y_{\text{obs.}}=100$ ) gives an additional score of verifications of the theory (though in the case of (II.) the observed "100" sets in somewhat too soon).

Grains in Clump		2	3	4	5	6	7	8	9	10	11	...	33
<i>a</i> .....		1.73	3.03	4.88	6.2	7.4	8.6	9.8	11	12	13	...	>25
I. ....	$\left\{ \begin{array}{l} y_{\text{calc.}} \\ y_{\text{obs.}} \end{array} \right.$	28.2 21	44.2 57	64.3 63.8	74.3 74.5	81.0 87.5	85.9 96	89.7 97	92.4 97	94.2 96.5	95.5 100	...	99.8 100
II. ....	$\left\{ \begin{array}{l} y_{\text{calc.}} \\ y_{\text{obs.}} \end{array} \right.$	15.3 13	25.3 37.6	41.2 42.3	49.3 53	56.4 66	62.6 82.5	67.9 86.5	73.5 (?)	75.9 89.4	78.8 100	...	95.7 100

The agreement, although in general not so close as in the previous case, is certainly satisfactory and in three or four instances even remarkably good. Notice especially the case of four-grain clumps which show perfect agreement in all three exposures, the calculated and observed values in the original exposure (*cf.* first paper) having been 87.3 as against 87.1, and now 64.3 and 41.2 as against 63.8 and 42.3. Almost the same is true of the five-grained clumps. But in general the agreement is good enough throughout the array of clumps\*.

4. Notwithstanding the good agreement and the consistency of these three sets of results with regard to the values of  $n$  and  $\sigma$ , some critical remarks must now be made about the meaning of the latter parameter. It will be remembered from the first paper that  $\sigma$  or  $\pi\rho^2$  was originally introduced as the (average) "cross-section" of the light darts, and  $\rho$  as their equivalent semi-diameter, and the mathematical rôle of this finite diameter was fixed by assuming that a grain is made developable only when it is "fully" struck by a light

\* The outstanding discrepancies being attributed mainly to the uncertainty of the (average) sizes  $a$  of the clumps and perhaps also to disregarding the effect of the finite range of  $a$  within each class of clumps. How this finite breadth of the classes can be taken into account will be shown presently.



dart. This gave as the efficient area of a grain, instead of  $a = \pi r^2$ ,

$$a' = a[1 - \rho/r]^2.$$

Now, exactly the same formula would arise if we assumed that, no matter what the thickness of the light darts (and whether it is finite at all), a grain is made developable only when the *axis* of the dart hits it in a point not too near the edge of the target (grain), thus excluding from the total area a boundary zone of a certain breadth  $\rho$ . Such a condition is not altogether fantastic, and one might support it by imagining that if the grain is hit too near its edge, an electron is still ejected and a "centre" of reducibility is produced at the spot, but the wave of development, stopping dead at the edge, has not such a good chance to spread over the whole grain as when the centre is well within the target. If so, then the empirical principle that a grain is either not affected at all or is made developable entirely would require a qualification, viz., the exclusion of that boundary zone. This alternative, therefore, should and can still be tested. If it is supported by experiment, the original interpretation given to  $\rho$  or  $\sigma$  can be abandoned, since it certainly is not very satisfactory. Not that there is anything incredible in the light darts having a finite thickness and a cross section such as one-tenth  $\mu^2$ ; so far as we know, they may be trains of waves of even much larger transversal dimensions. But the unsatisfactory point about this interpretation is that it is hard to imagine why the grain to be affected at all, *i. e.* to have a photo-electron ejected, has to be hit by the whole of that cross section. For, if so, then, unless some light darts have a diameter of the order of  $10^{-8}$  cm., no such things as simple atoms or molecules could ever have their electrons ejected by light\*. Yet, a grain, as a crystal lattice, may, after all, behave as a single molecule, at least in the present connexion, and the original rôle attributed to the cross-section of the light darts, though repugnant, may still turn out to be a useful working hypothesis. To ensure the possibility of being fully hit and therefore affected, even to the smallest available silver halide grains, it would be enough to treat  $\sigma$  in our formula as the average taken over a sufficiently ample interval of sections down to very small ones. It would be premature to enter into quantitative details of the consequences of such an assumption. But it seems proper to mention even at this stage that an assumption

\* Whereas the photo-electric effect has been obtained with gaseous substances, though not beyond every doubt.

of this kind can well be tested experimentally. In fact, if that assumption be correct, then the light traversing two or more equal photographic plates piled upon each other should contain, successively, a larger percentage of the coarser light darts, so that the formulæ of type (12) representing the number of affected grains or clumps of various sizes should have not only a decreasing  $n$ , but also a successively increasing average value  $\sigma$  of the cross-sections of the darts, a comparatively larger proportion of the more slender darts being absorbed each time. In short, we should have a kind of sifting effect. Such experiments which, to be at all convincing, require obviously a much higher degree of accuracy in counts and area measurements, are now in preparation. Their results will be published in a subsequent paper. In the meantime, the parameter  $\sigma$  may and profitably will be retained as a small but desirable correction of the exponential formula without, however, being given either of the alternative interpretations.

It may be well to add here also a few remarks about  $n$ , the chief parameter in the fundamental formula. This was originally defined as the number of light-quanta or darts thrown upon the photographic plate per unit of its area. Now, apart from the generally small correction term containing  $\sigma$ , the parameter  $n$  appears in the formula only through the product

$$p = na,$$

where  $a$  is the area of the grain. Thus, essentially only the value of this product (a pure number) can be determined from microphotographic experiments. Suppose now that the sizes of all grains of the given emulsion were reduced in the same ratio, converting every  $a$  into  $\epsilon a$ ; then, the same experimental value of  $p$  would indicate a number of light darts  $\frac{1}{\epsilon}$  times larger. Now, such would exactly be the position if for every grain not the whole but only a fraction  $\epsilon$  of the area were vulnerable, *i. e.* deprived of an electron on being hit by a light dart. The grain may be sensitive only in spots scattered over its area, and each perhaps of very minute dimensions. Provided that all these spots occupy a *fixed* fraction  $\epsilon$  of the total area of the grain, the microphotographic counts and measurements could not inform us about the value of this fraction unless the exposure given to the plate is known in absolute energy measure. Thus, for instance, if, as was tacitly assumed,  $\epsilon = 1$ , the number of light darts in the set (I.) of observations just described would lead

to  $n=0.286$  per  $\mu^2$  or about 29 million darts per square centimetre of the plate; but if, say, only one-thousandth of the area of each grain were vulnerable, we should conclude that 29 milliards of darts were thrown upon each  $\text{cm.}^2$  of the plate. But it would be idle to speculate upon this subject and, as far as we can see, the only way of deciding whether that suggestion is correct or not and of determining the value of the fraction  $\epsilon$  is to measure the exposure energy in absolute units\*. Now, in none of our experiments thus far reported was the energy value of the exposure even roughly estimated, not to say measured. But in order to decide this important question, preparations for measurements of this kind are now in progress in this laboratory, and their results will be published in due time.

5. *Effect of finite breadth of size-classes of targets.*—The short name "target" will now be used for either a single grain or a clump of grains in sufficient contact to act as a photographic unit.

In the three sets of observations hitherto reported, the targets were classified according to the number of grains contained in them (from 1 to 33), and for each class the average size (area) was used as  $a$  in the theoretical formula, without taking account of the finite breadth of any such class, *i. e.* of the interval,  $a_1$  to  $a_2$  say, over which its individuals ranged. It was not possible with the said classification to secure reliable estimates of this breadth, which, however, for some classes might have been considerable (perhaps of the order  $1\mu^2$ ), and at any rate varied from class to class. It is likely that some of the outstanding discrepancies are due to these neglected factors and especially to the latter.

To eliminate this source of error, and at the same time to avoid the laborious planimetrization of targets within very narrow limits, we propose henceforth to divide the whole material of targets into deliberately broad classes, all of equal breadth, say  $2\alpha$ .

If, then, the average size of any of these classes of targets is used as the variable  $a$  in our formula, a correction has to be made for the finite value of  $2\alpha$ . This correction can easily be found.

Disregarding for the moment the  $\sigma$ -term, the number of targets of a class of breadth  $2\alpha=a_2-a_1$  affected by  $n$  darts, is

\* Although even then the final result would be made doubtful by the uncertainty whether the total light energy (as required by Einstein) or only a fraction of it is conveyed in discrete quantum parcels.

by the fundamental formula (7), first paper,

$$k = \int_{a_1}^{a_2} f(a) [1 - e^{-na}] da,$$

where  $f(a)da$  is the number of targets of size  $a$  to  $a+da$  originally present. Now, if  $2\alpha$  is of the order of  $1/2$  or even  $1\mu^2$ , we can take  $f(a) = \text{const.}$  within the integration interval with sufficient accuracy for all of our experimental emulsions. Thus, denoting by  $N$  the original number of targets in the whole class, so that

$$f = f(\bar{a}) = \frac{N}{2\alpha},$$

we shall have 
$$\frac{k}{N} = 1 - \frac{e^{-na_1} - e^{-na_2}}{2n\alpha},$$

or, writing simply  $a$  for the average  $\bar{a} = \frac{1}{2}(a_1 + a_2)$ , and therefore,  $a_2 = a + \alpha$ ,  $a_1 = a - \alpha$ ,

$$\frac{k}{N} = 1 - e^{-na} \cdot \frac{e^{n\alpha} - e^{-n\alpha}}{2n\alpha}$$

Remembering that  $\frac{1}{2}(e^{n\alpha} - e^{-n\alpha}) = \sinh(n\alpha)$ , writing for brevity

$$v = \log \frac{N}{N-k}, \quad . \quad . \quad . \quad . \quad . \quad (13)$$

and replacing  $a$  in the chief term by

$$a' = a[1 - \sqrt{\sigma/a}]^{2*},$$

we have ultimately the required formula

$$v = na' - \log \frac{\sinh(n\alpha)}{n\alpha} \quad . \quad . \quad . \quad . \quad (14)$$

Notice that the correction term depends only on  $n\alpha$ , that is to say, for  $\lambda = \text{const.}$ , on the product of the exposure and the class breadth. If this product is a fraction, such as one-half or even two-thirds †, we can write, up to  $(n\alpha)^4$ ,

$$v = na' - \frac{1}{6}(n\alpha)^2 \quad . \quad . \quad . \quad . \quad (14a)$$

If, as explained, all the contemplated targets of the emulsion are divided into classes of *equal breadth*  $2\alpha$ , the

\* This is accurate enough provided  $\sigma/a$  is small. In the correction term the semi-breadth  $a$  requires practically no amendment.

† If  $\alpha = 1\mu^2$  and the exposure is as in the previous concrete cases, the value of this product does not exceed 0.6.

correction term in (14) is, for a given exposure, constant throughout the array of classes, and  $v$  plotted against  $a$  should give a straight line. If  $\sigma$  were non-existent or negligible, we should have a straight line for  $v$  plotted against  $a$  itself.

The aforesaid classification of targets and the corresponding formula (14) will be used for analysing all the experiments now in progress. For the present, we are able to quote only one such set of results condensed in the following table.

The targets (grains and clumps alike) were all divided into five classes of equal breadth  $2a = 0.60\mu^2$ , ranging from 0.20 to 0.80, from 0.80 to 1.40, etc., as shown in the first column, which gives the average sizes  $a$  in square microns. The third column gives the observed number of targets surviving for every  $N$  targets originally present, each of these data being an average of counts on four different domains of the plate. The fourth column contains the percentage number  $y = 100 \frac{k}{N}$  of grains affected, as observed, and the fifth, as calculated by (14), to wit, with

$$n = 0.255 \text{ per } \mu^2,$$

$$\sigma = 0.0081\mu^2.$$

$a$ .	$N$ .	$N-k$ .	$y$ obs.	$y$ calc.	$\Delta$ .
0.50	190.3	173.3	8.9	9.0	-0.1
1.10	140.0	103.5	26.0	(20.6)	(+5.4)
1.70	62.6	43.0	31.3	31.1	+0.2
2.30	31.4	18.7	40.4	40.3	+0.1
2.90	19.8	10.3	48.0	48.4	-0.4

The agreement is, apart from the second class, bracketed as an "outlaw," almost perfect. The "cross-section" of the darts, or what  $\sigma$  may stand for, is agreeably about ten times smaller, *i. e.* the diameter three times smaller than that previously obtained with the same light source. This is not to say that the reality and rôle of  $\sigma$  is herewith settled. Yet it is interesting that without  $\sigma$ , that is to say, with  $a'$  in (14) replaced by  $a$  itself, no choice of  $n$  yields such a close agreement. As to the correction term due to the finite class breadth, it may be mentioned that in the present case it amounts (as a subtrahendum from  $v$ ) only to 0.030.

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