

THE UNIVERSITY OF MICHIGAN
COLLEGE OF LITERATURE, SCIENCE, AND THE ARTS
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PART II. THE PHOTOGRAPHIC SEQUENCE EFFECT

J. H. Enns
Ernst Katz
Project Supervisor

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PREFACE

The study of the photographic latent image, reported in Part II of this final report, started in 1952 under Contract DA-20-018-ORD-12258 with the Office of Ordnance Research of the U. S. Army. To simplify administration, it was merged at the request of the principal investigator into Contract AF 18 (600)-750 with the Office of Scientific Research of the U. S. Air Force in 1955. The latter was reorganized into the present Contract AF-49(638)-69 with the same office in 1956.

The study of the photographic latent image led to the Ph.D. thesis of Dr. R. L. Martin³ in 1956. A paper was published by Dr. J. H. Enns and Dr. E. Katz.² The experiments described in the present report were carried out by Dr. Enns. A paper on the material covered in the present report will be submitted for publication in the future. At various scientific meetings,^{4,5} four papers were read to communicate results of our research on the photographic latent image.

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ABSTRACTS

PART I. GALVANO-MAGNETIC EFFECTS

A historical preface is given. In Section 2 the phenomenological theory as developed by Kao and Katz is summarized, with special emphasis on applications to bismuth and gallium. In Section 3 the experimental method of measurement and results for bismuth are described. It is found that all resistivity brackets can be represented by the formula $\{ \quad \} = \Gamma_0 T e^{-\beta \sqrt{T}}$, where Γ_0 is a constant different for each bracket and β depends almost exclusively on the order of the bracket. This dependence is roughly given by $\beta_n = 2n/(n+4)$. No theoretical interpretation of these results is presently available. Their compatibility with current models of the energy surfaces of electrons and holes in bismuth is being tested. The amount of labor involved in this testing has precluded our reaching a definite conclusion at the time of this writing. The question will be pursued further. In Section 4 results from the literature for gallium are analyzed in terms of our phenomenological constants. Here also relations of the above form hold. Again the electron theoretical implications remain to be worked out. In Section 5 the procedure of testing and its complications are sketched, as is the direction in which an entire new series of bracket relations is in process of being discovered by the present analysis.

PART II. THE PHOTOGRAPHIC SEQUENCE EFFECT

In Section 1 the motivation of this study is given: comparison of limiting low-intensity reciprocity failure from sequence loops with direct measurements by Martin, and study of the number of quanta active in the formation of a just developable latent image as a function of grain size, density, and type of development by means of sequence loops. In Section 2 the method of sequence exposures is briefly described experimentally and theoretically and the experimental results are described. These were obtained with Eastman Kodak type 33 emulsions and with a set of emulsions, kindly supplied especially for this study by Eastman Kodak laboratories, consisting of nonsensitized pure AgBr emulsions of different average grain sizes. The results confirm qualitatively the trend of the theoretical work but lead to quantitative difficulties, which are discussed in Section 3. The conclusions are summarized in Section 4. At present no practical way is seen to refine the theory to a degree that would make for its quantitative applicability. To do so would require introducing too large a number of unknown parameters and make it very complex. For qualitative use the theory and the method of sequence-loop studies seem useful tools for the determination of the number of quanta that are involved in the formation of one just developable speck. Values in the range 3 to 20 quanta were found in the present studies. This number tends to be lower for grains that require a larger total number of quanta received to become developable, indicating that the efficiency of concentrating the received energy on one speck tends to be lower for grains with smaller just developable specks. No explanation of this trend is presently available.

PART II. THE PHOTOGRAPHIC SEQUENCE EFFECT

1. Introduction

In 1950 E. Katz¹ published a paper which opened the possibility of quantitatively analyzing under certain conditions the sequence effect, i.e., the difference in efficiency of producing a developable image by a high-intensity exposure followed by a low-intensity exposure as compared to that of the same exposures in reverse order. If both intensities are well below the optimum intensity of the particular emulsion, the study of the sequence effect permits evaluation, according to this theory, of mainly two parameters that cannot easily be evaluated otherwise. The first is the "limiting slope" of the reciprocity failure diagram. It refers to the slope of a log exposure versus log intensity plot for constant density in the limit of two intensities. The second is the minimum number of quanta absorbed by the average grain to become developable. Both parameters furnish information about the process of latent image formation.

Apparatus was built to expose plates in the manner required to various exposures of two intensities differing by a factor of 100. The apparatus and method were tried out on an Eastman Kodak Type 33 emulsion, and the results of this work, which were published,² looked reasonable and promising; consequently, it was decided to study the results of this method for a number of emulsions which had been kindly prepared specially for us by the Eastman Kodak Co.* These were pure AgBr emulsions with different known grain sizes and minimum sulphur sensitization. They were to be studied both with internal and with surface image development. The data thus aimed at would provide a check on the general applicability of the method. They also would correlate with the results of the thesis of R. L. Martin³ who studied their low-intensity reciprocity failure by a direct sensitometrical method.

The results obtained from these emulsions are described in Section 2. The description follows essentially the same lines as a paper⁴ presented at the Chicago Meeting of the American Physical Society in March, 1958. The data are interpreted according to the lines of thought laid down in the 1957 paper.²

The period after March, 1958, was spent principally in trying to overcome certain difficulties in interpretation which had meanwhile become apparent to us. These difficulties are briefly described in Section 3. They turned out to be more fundamental than was at first apparent. As a result, we presently

* The cooperation of the staff of the Eastman Kodak Co. Research Laboratory in making these plates available and also in helpful discussions is gratefully acknowledged.

regard the conclusions drawn in Section 2 as preliminary, and probably correct only in a semiquantitative sense. However, it has also become clear that really quantitative interpretation of our results would first require a great deal of further theoretical work, which could not be accomplished in the available time. The situation is briefly sketched in the concluding Section 4.

The entire effort should be seen against the background of the present status of photographic latent image theory. It represents one of the efforts to make latent image effects subject to quantitative measurement. While much is known qualitatively, very few of the simplest basic effects in this field are amenable to experimentation that allows quantitative interpretation in terms of basic theory. The present work looked like a possibility in its initial stages, but turned out to be subject to more complicating factors than were anticipated. While it may be possible in the future to develop the present method further, the conclusion at present must be that the effects studied do not lend themselves directly to the quantitative purpose for which the study was undertaken.

2. Photographic Emulsion Studies by the Sequence-Exposure Experiment

2.1. INTRODUCTION

In general, any experiment designed to yield exposure data of the individual photographic grain is complicated by the uncertainties of the effective grain size and the effective light intensity incident on a grain. In such experiments it is common to work with single-grain-layer emulsions. The grain size and the effects of exposure are then derived from microscopic grain counts of large numbers of undeveloped and developed grains. The effective light intensity is that appearing at the front surface of the emulsion.

In the photographic-sequence-exposure experiment, whose basic principles we have described in a publication,² one does not require an explicit value for the grain size nor the light intensity incident on a grain. Consequently, emulsions of standard thickness can be studied and all densitometry readings are made in the usual way. The experiment is based on the hypothesis that, for isodense exposures, the probability of rendering the last grains developable must be equal. In the development of the theory it is shown how one can obtain information from isodense sequence loops concerning the number of photons absorbed by the just developable grain, and also the fraction of these required to transform a stable system into a developable latent image. Let us briefly consider the main points of the experiment and related theory.

2.2. EXPERIMENTAL METHOD

In a sequence exposure the emulsion is given an exposure I_1T_1 followed by an exposure I_2T_2 . The total exposure is then the sum $E = I_1T_1 + I_2T_2 = pE + qE$, where $p + q = 1$. For sufficiently large intensity ratios, say $I_1/I_2 = 100$ to 1000 , and both intensities below the optimum value, the result-

ing density is one function of E and p for the case when the high-intensity exposure is followed by the low-intensity exposure, and a different function when this order is reversed. This is illustrated by the isodense loops shown in Fig. 1. Here $\log E$ has been plotted as a function of p , where p denotes the ratio of the first exposure to the total. The upper curve is the L + H exposure (low-intensity followed by high-intensity), while the lower curve represents the H + L data. In this particular case the intensity ratio was $I_H/I_L = 160$. In most of the results presently reported I_H/I_L was exactly 100, unless stated otherwise.

2.3. THEORY

As previously mentioned, the theory of isodense exposures leads to the statement that the probability for rendering the last grains developable must be a constant. This can be expressed in equation form as

$$cWN = \text{constant.} \quad (1)$$

c is considered to be a constant, independent of the light intensity, and is the chance that two simultaneously present photoelectrons will combine at the same trap to form a stable speck. Its value, between 0 and 1, is not known. However, since c is taken to be independent of the light intensity, it will cancel from the equations that follow.

W represents the probability that a second photoelectron will be liberated while the first is still "alive." Its dependence on the intensity I , the exposure time T , and the limiting slope of the low-intensity reciprocity failure is given by the equation

$$W = \int_0^T F(t) P(t) dt, \quad (2)$$

where $F(t)$ is the probability of survival of a photoelectron at time t after its generation and $P(t) dt$ is the probability of a quantum generating a second photoelectron in the interval between t and $t + dt$ after the first was generated.

N in the original expression is the number of interquantum times occurring during the formation of the stable speck. For a just developable grain having been subjected to an exposure, rated at E photons, N would be, according to the theory published

$$N = E - 1 + e^{-E} - E_0, \quad (3)$$

where E_0 represents the number of photons that were absorbed to transform the stable image to the just developable state.

The general expression relating isodense exposures is given, according to the published article by

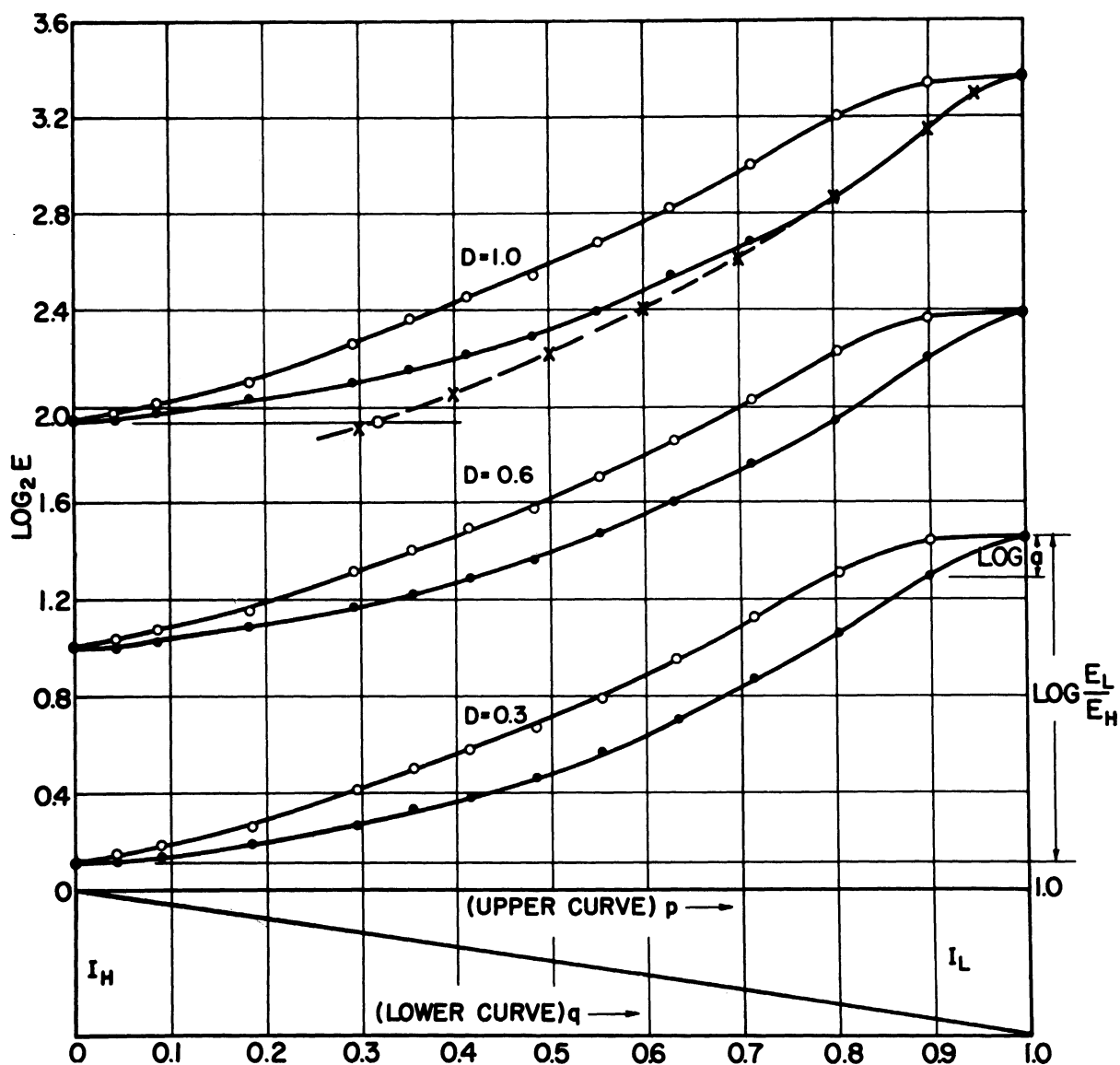


Fig. 1. Typical set of isodense loops from one plate for densities 0.3, 0.6, and 1.0. Upper curves: low-intensity followed by high-intensity (L+H); lower curves: reverse order (H+L).

$$W_1 N_1 + W_2 N_2 = W_L N_L \quad (4)$$

or with (3)

$$W_1 (pE - 1 + e^{-pE}) + W_2 (qE - 1 + e^{-qE} - E_0) = W_L (E_L - 1 + e^{-E_L} - E_0) \quad (5)$$

Here the sum of the probabilities to form a stable speck as a result of the two sequence exposures on the left is equated with the corresponding probability of the isodense terminal exposure of the loop on the right, which was made all at low intensity I_L .

Further progress requires an explicit form for W . According to Eq. 2 the form of $P(t)$ and $F(t)$ is required. The form of $P(t)$ is well known from statistical considerations of light absorption:

$$P(t) = (1 - \frac{t}{T}) e^{-It} \quad (6)$$

The form of $F(t)$, the "survival function", is not as certain. However, we have shown previously that to arrive at a reciprocity failure curve which would be straight with slope $-\alpha$ for low intensities, there is little choice and the form of F must be essentially

$$F(t) \approx \left(\frac{1}{\lambda t}\right)^\alpha \quad (7)$$

Substitution of (6) and (7) into (2) leads to

$$W = (\lambda T)^{-\alpha} e^{-It} + (1 - \beta/IT) (I/\lambda)^\alpha \Gamma(\beta) \quad (8)$$

where $\beta = 1 - \alpha$. Substituting this into (5) with appropriate indices for the first and second part of the exposure gives the equations for the upper as well as the lower branch of the loop. A valuable approximate simplified form, which is valid when neither p nor q is very small, was used for the interpretation of the lower H+L branch:

$$E^2 [w p^2 + p(q - a)] - E w p (1 + \beta) + \beta [w + p(1 + E_0) \left(\frac{1}{q} - \frac{1}{a}\right)] = 0 \quad (9)$$

where $w = (I_H/I_L)^\alpha$ and $a = E_L/E$. A similar expression can be derived for the upper L+H branch.

With the help of these equations and the exposure ratios available from experimental loop data, it is possible to evaluate E , E_0 , and α . It is recalled that E is the number of quanta absorbed, each producing a photoelectron, required to make a developable latent image speck in the last grains rendered developable; E_0 is the number of quanta (photoelectrons) required to bring the stable sublatent image to developability; α is the limiting slope of the low-intensity-reciprocity-failure curve.

2.4. EXPERIMENTAL RESULTS

Figure 2 shows results obtained for two plates of Eastman Kodak emulsion type 33. The number of photons absorbed to produce a just developable grain at the higher of the two intensities, E_H , is shown as a function of density. The two plates differed considerably in age (about two years) and hence in fog density D_f . The increase in E_H with density is qualitatively attributed to reciprocity failure. For at higher densities the grains becoming developable last are smaller and lie deeper in the emulsion; hence their rate of quanta reception is lower and they react with a lower efficiency corresponding to a lower intensity. Also the lower E_H curve for the older plate with more fog can be accounted for qualitatively on the same basis; one must then assume that aging these emulsions has an effect roughly analogous to a pre-exposure, with the distinction, however, that the smaller or less sensitive grains age more than the larger, more sensitive ones. In this connection it is interesting to note that the curves converge for $D \rightarrow 0$ to $E_H \approx 5$ or 6 . This value for E_H for the most sensitive grains of the emulsion appears to be essentially independent of aging. We have not obtained microscopic evidence to support the above interpretation, such as might be derived from a study of the grain sizes in the fog on both plates.

The intensity at which the above data were taken was equivalent to $I_H = 1.25$ quanta per average grain per second and $I_L = I_H/100$. The optimum intensity of this emulsion is approximately ten times this value.

Figure 3 shows curves of E_H'/E_H versus density for the same two plates. E_H' is the front surface incident exposure for the average grain size. The two curves almost coincide, showing that this ratio is not sensitive to aging, which is accompanied by a decrease in reciprocity failure for this emulsion type. Also, convergence to unity at zero density is interpreted as meaning that these grains are essentially perfect absorbers of the 4358\AA radiation used in these experiments, in conformity with general evidence from the literature. It is remarkable that the sequence-exposure experiments permit such conclusions for which normally quite different types of experiments would be required.

Figures 4 and 5 show data of the same sort as Figs. 2 and 3, taken with special nonsensitized pure silver bromide emulsions of different grain sizes, specially prepared by the Eastman Kodak Co. The figures show two sets of results with different prestorage temperatures. The plates were stored in a refrigerator but were prestored for 24 hours immediately before the sequence exposures at temperatures of 25°C or 50°C as indicated.

The most important result here is that, for prestorage at 25°C , the curves slope downward to the right, in contrast to Fig. 2. For prestorage at 50°C , the slope is less for corresponding grain sizes. The decreasing slope with higher prestorage temperature seems analogous to the decreasing slope with aging observed in Fig. 2 for type 33 emulsions. Also, the common intercept for $D \rightarrow 0$ is an analogous feature. However, in contrast to the results for type 33,

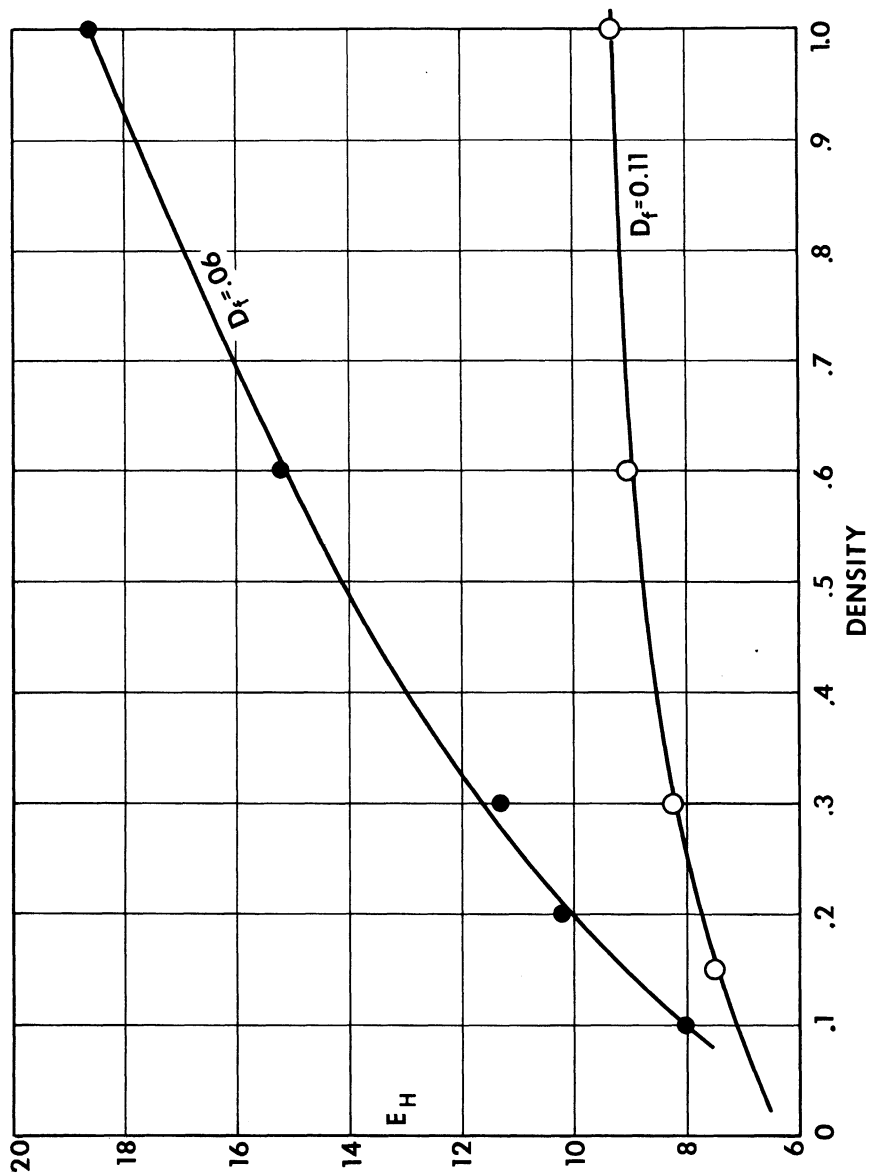


Fig. 2. Number E_H of photons absorbed to produce a just developable grain, as a function of density D . Eastman Kodak type 33 emulsion, intensity $I_H = 1.25$ quanta per average grain per second, fog density D_f according to age, 4358\AA radiation.

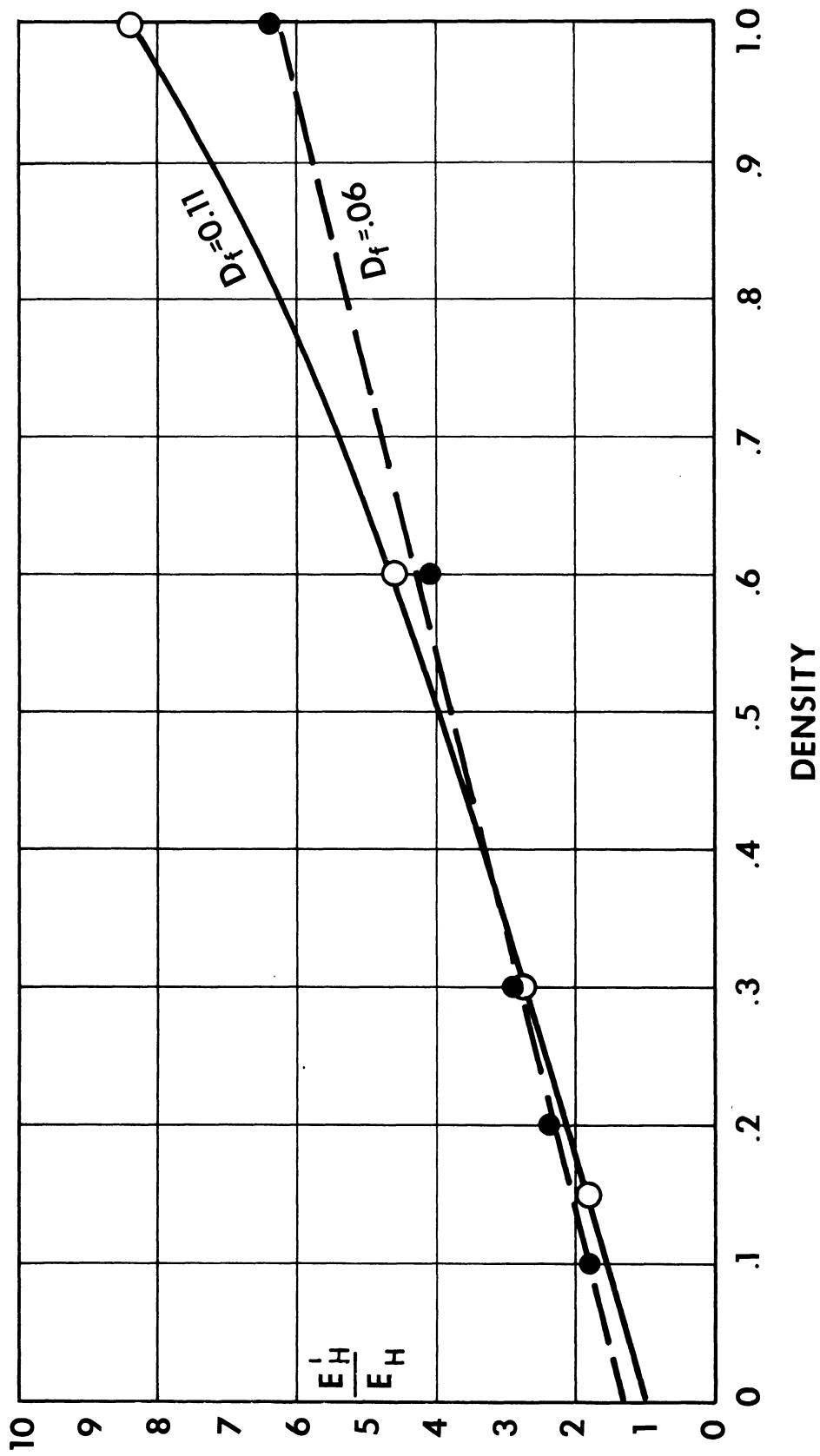


Fig. 3. Ratio of front surface incident exposure per average grain E_H' to E_H for the plates of Fig. 2.

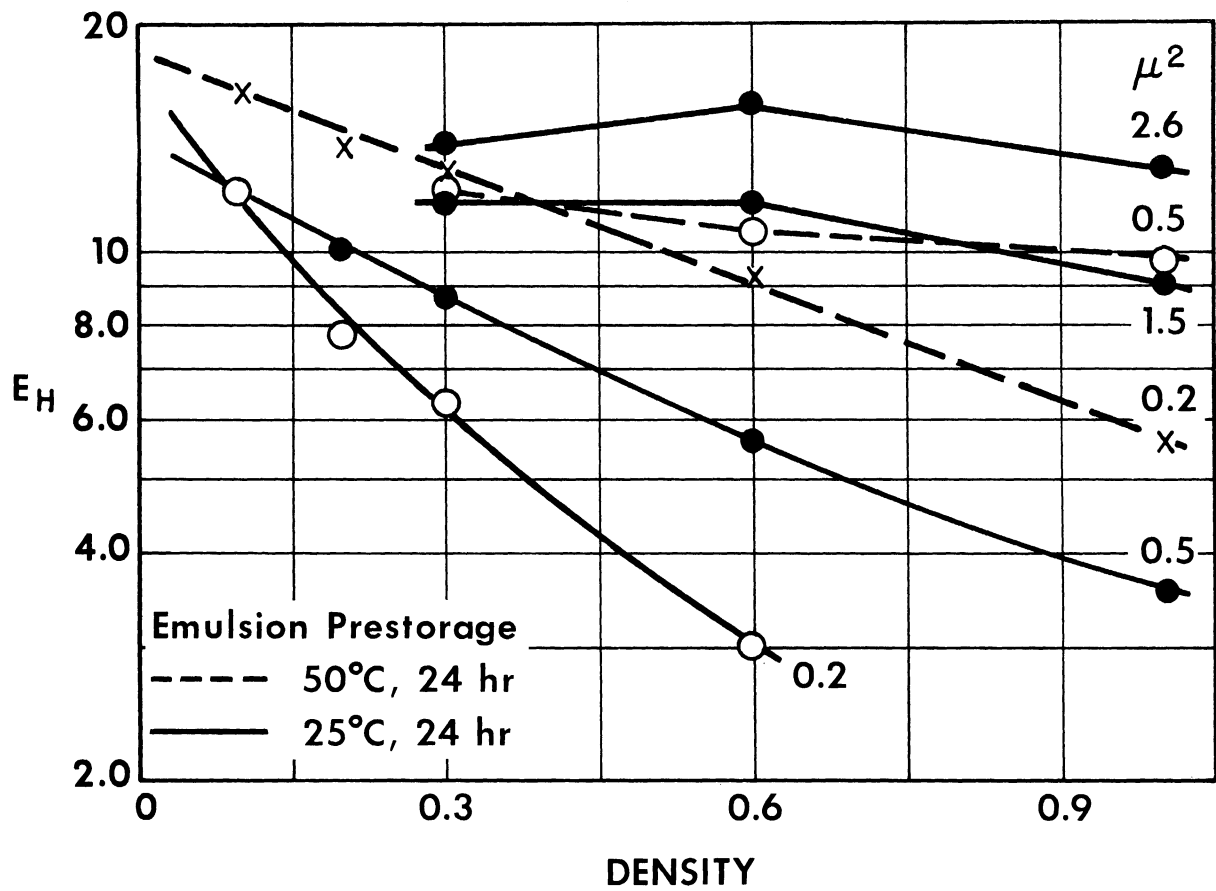


Fig. 4. E_H versus density as in Fig. 2 for special nonsensitized pure silver bromide emulsions of different grain sizes. Average cross-sectional area of grains in square microns as marked.

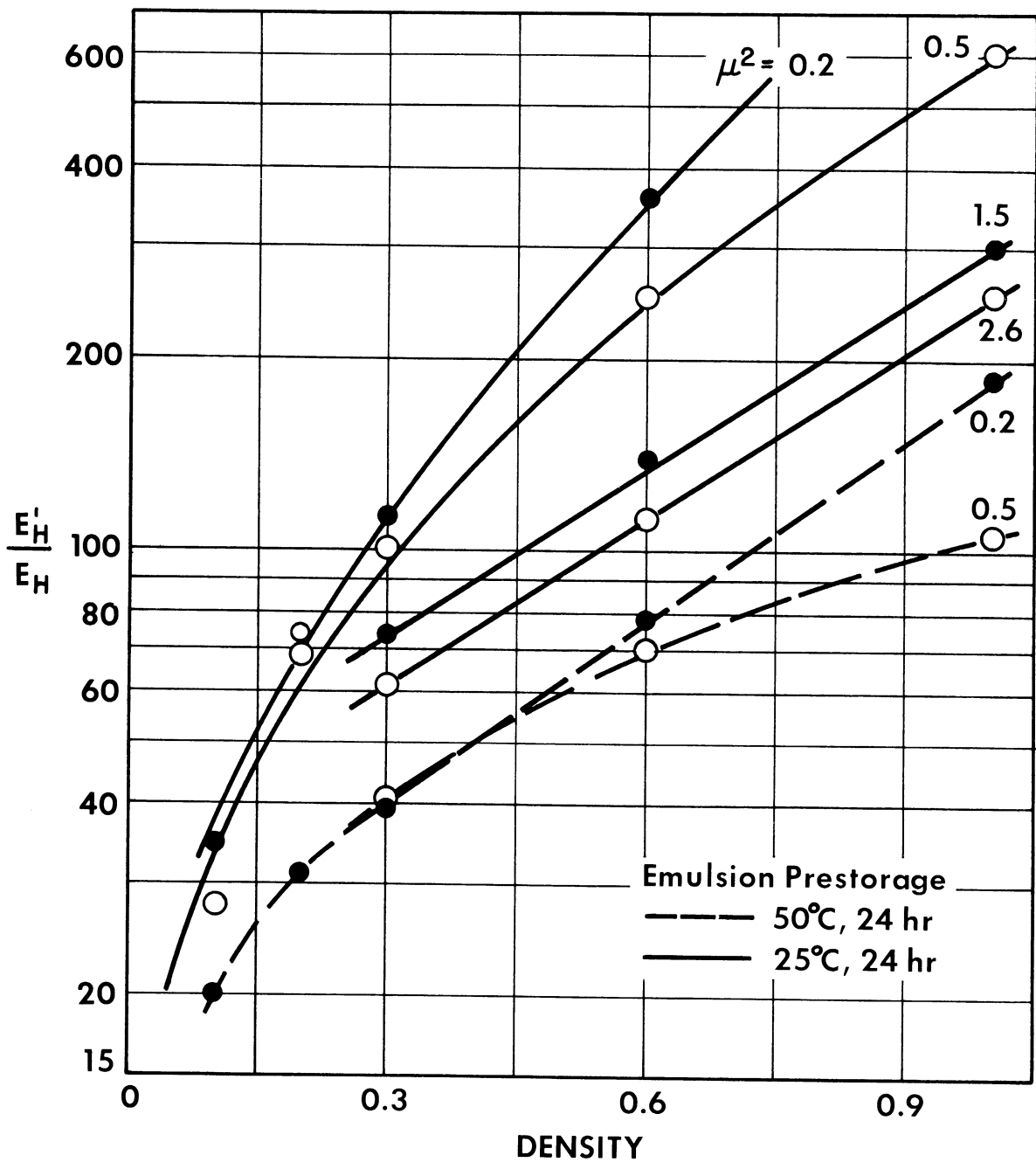


Fig. 5. E_H'/E_H versus density as in Fig. 3 for special nonsensitized pure silver bromide emulsions of different grain sizes. Average cross-sectional area of grains in square microns as marked.

the pure silver bromide emulsions show that the less sensitive grains, i.e., those that become developable relatively late, require fewer absorbed quanta for developability. This feature is manifest in three ways:

- (a) For each curve, the grains at higher density, which are less sensitive, have lower E_H values.
- (b) Comparing curves for different grain sizes, the smaller grains, which are on that account less sensitive to a given incident intensity per unit area, show lower E_H values.
- (c) The plates prestored at 25°C are less sensitive than those prestored at 50°C and the former show lower E_H values.

It is not clear, even qualitatively, how to explain this trend. A possible explanation may be related to a greater degree of latent image dispersion (the formation of more stable sublatent image specks) in the more sensitive grains, but a good deal more experimentation would be required to confirm such a hypothesis. The conclusion, while supported independently, by three variables, hinges on the correctness of the analysis of the experimental curves from which they are derived by interpreting them in a certain way. If this interpretation were to be modified, a new set of curves would result to replace those of Fig. 4. Such modification would seem imminent in view of the extremely low values obtained for E_H on some curves. The curve for 25°C prestorage and $0.2\mu^2$ average grain area has a measured point at $E_H = 3$, and that of $0.5\mu^2$, at $E_H = 3.6$, and the general trend of the curves suggests that they may well go below $E_H = 3$. This point will be taken up in Section 3 as one of the major difficulties encountered in this work.

Figure 5 shows the ratio of incident to absorbed photons E_H'/E_H for the same emulsions, as a function of density. The ratios are many times larger than those for type 33 emulsions, indicating that pure nonsensitized silver bromide emulsions are not as efficient absorbers of the 4358Å radiation and/or less efficient converters of absorbed quanta to photoelectrons. This effect is more pronounced with increasing density, decreasing grain size, and lower prestorage temperature. These data merely lend increasing support to the conclusion drawn from Fig. 4. The grains which are the least sensitive in these emulsions, even when expressed in terms of quanta received per grain area, are apparently inefficient in some part of the process of latent image formation which lies before the actual formation of the silver speck. The formation of the silver speck itself is particularly efficient with those grains that are inefficient in the other parts of the process.

Figure 6 shows the limiting slope of the low-intensity reciprocity failure as derived from the same experimental data. This figure is qualitatively similar to Fig. 5 and qualitatively an inverted image of Fig. 4, indicating that large reciprocity failure means low overall efficiency of the latent image formation but high efficiency in that part of the process which refers to the formation of the silver speck. It is typical that, in the same range where the

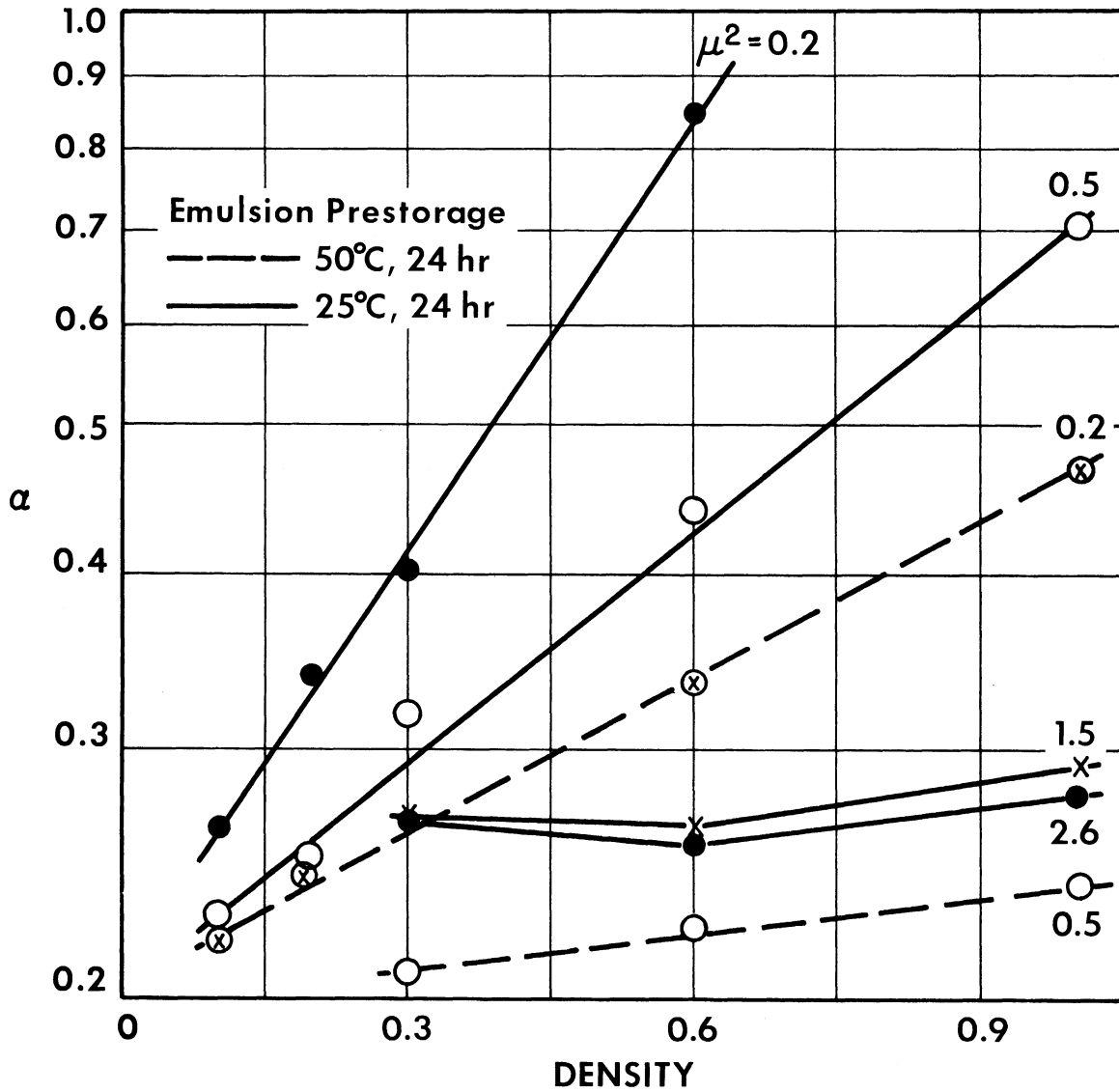


Fig. 6. Limiting slope α of low-intensity reciprocity failure derived from sequence loop data for special nonsensitized pure silver bromide emulsions of different grain sizes. Average cross-sectional area of grains in square microns as marked.

curves of Fig. 4 threaten to go below the value 3 which indicates that something is wrong (see Section 3), the curves of Fig. 6 threaten to go above $\alpha = 1$, which would also be in conflict with the theory used. However, remarkably enough, no points were obtained at this stage with $E_H < 3$ or $\alpha > 1$, since the exposure times required for these small grain emulsions and high densities fell beyond our program and would have required a special effort which was not undertaken at the time. One cannot, therefore, exclude the possibility that the curves bend rather sharply at these values and actually remain above $E_H = 3$ and below $\alpha = 1$.

Figure 7 gives curves for E_H'/E_H versus grain area for the various densities for two types of development. The negative slope of the solid lines representing total image development (the same development as for all previous figures) is consistent with the relative location of the curves for the densities 0.3, 0.6, and 1.0, i.e., to increase the grain area has the same qualitative effect on E_H'/E_H as to decrease the density. The broken curves are for development of the internal latent image only. The conditions of development of the internal image are the same as those used by Martin.³ The internal image curves show that E_H'/E_H increases for increasing grain size and for increasing density. These two features of the internal image seem to be in conflict with each other.

A quantitative comparison of limiting slopes α between Martin's and the present work is not feasible because the curves of Martin indicated that they had not yet reached low enough intensities. They would require extrapolation. Also, the sensitivity of the limiting slope as found in the present work with respect to prestorage conditions makes quantitative comparison difficult. In a general qualitative way, the results presented in Fig. 6 are certainly compatible with those of Martin.

3. Difficulties

As previously indicated, a number of difficulties arose in the process of interpretation of the data. These difficulties did not stem from inaccuracies in the experimental procedure, which was carried out with meticulous care by Dr. Enns and showed a most satisfactory degree of reproducibility.

3.1. THE REQUIREMENT $E_H \geq 3$

The existence of the reciprocity failure shows that a stable sublatent image speck consists of at least two atoms; hence its formation requires at least two quanta. The sublatent image is not the same as the developable latent image; hence at least one more quantum is required to produce the developable latent image. Thus at least three quanta are required to produce a developable speck or $E_H \geq 3$. The fact that the curves of Fig. 4 tend below $E_H = 3$ can be considered a difficulty. It may be argued that this is man-

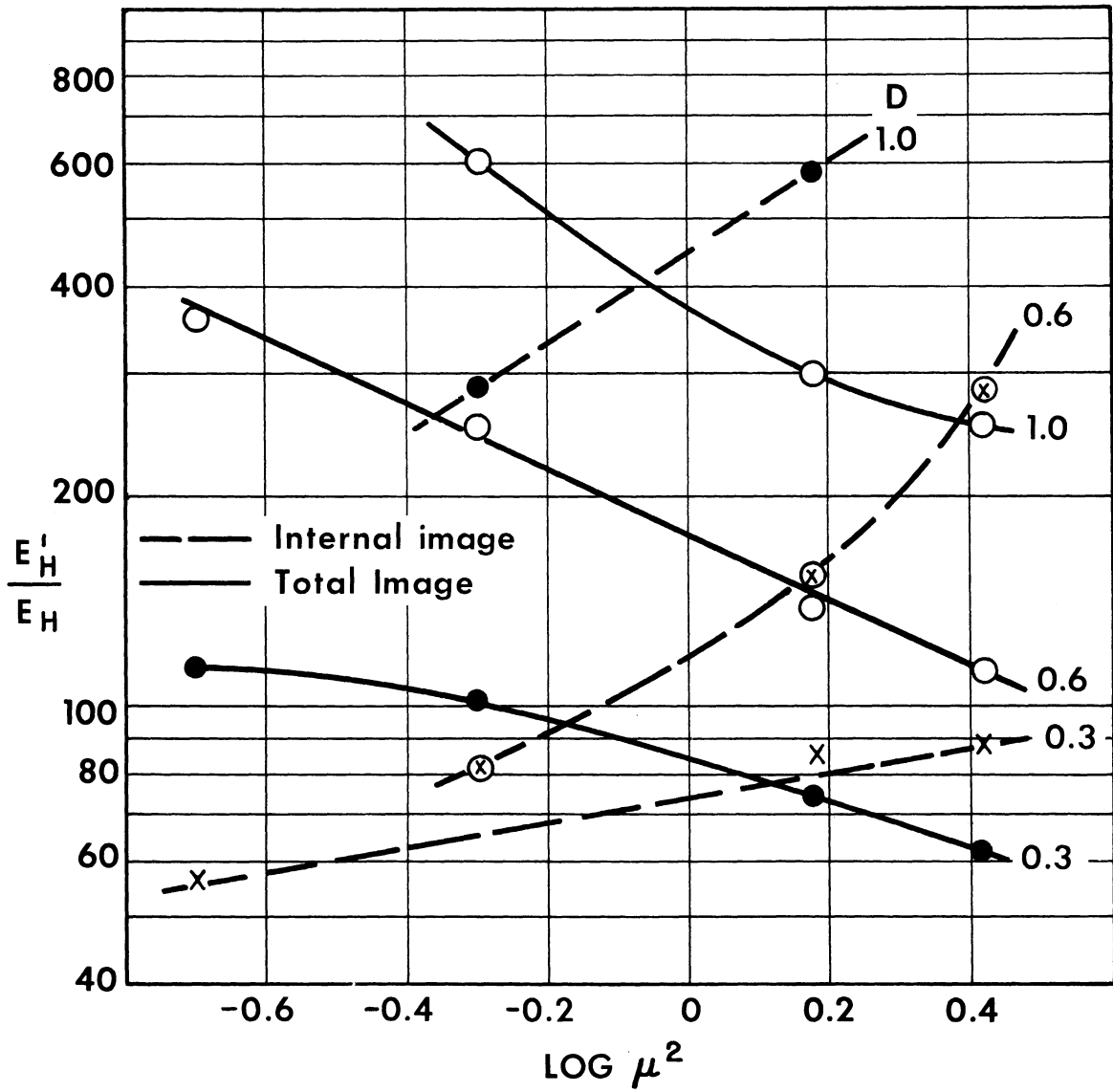


Fig. 7. E'_H/E_H versus grain area in square microns for various densities for two types of development. Special nonsensitized pure silver bromide emulsions.

ifest only through one point. At this stage it is a serious warning but not yet an outright conflict. See, however, Section 3.2 below.

3.2. AN ERROR IN REFERENCE 2

Scrutiny of the theory as presented in Ref. 2 and in Section 2 of this report shows that the theory contains an error of a rather subtle nature in the form of the quantity W . If this error is corrected, the net result is that all values of E_H reported so far, including those of Section 2 of this report, must be reduced by one. This fact in connection with the requirement of Section 3.1 represents now an unambiguous conflict.

3.3. A DIFFICULTY IN THE ANALYSIS OF THE SEQUENCE LOOPS

In Ref. 2 it is explained that the analysis of the loop for the extraction of the data presented involves plotting the reciprocal slope of the upper curve as a function of the exposure fraction p . Theory expects a straight line of slope minus one for this plot, at least for a substantial intermediate range of p values. This was not often the case. Frequently no straight section was discernible, or the slope was far from minus one.

The fact that the reciprocal slope plot rises above the expected straight line for values of p above 0.5 or 0.6 is not surprising and has to do with the theoretically expected gradual bending over of the upper curve of the loop as one approaches the right terminal. This is not part of the difficulty in principle but only in practice, if the influence extends too far down to lower p values. In principle one could correct for this if one knew a good deal more about the distribution of grain sizes.

The fact that the reciprocal slope plot rises above the straight line for low values of p , 0.3 or so, is more serious. The origin of this deviation escaped qualitative interpretation for a long time. It was finally traced as being due to the following experimental detail. The theory assumes that the two successive exposures are separated by a long interval of no light. The data of the lower curve were taken in agreement with this assumption. However, technical expediency had led to a programming cycle which would expose the plate to two exposures in immediate succession for the upper curve. This seemingly trivial point turned out to be responsible for the deviations. A correction was estimated for one curve, which made it come straight. However, the general form of the correction was prohibitively complicated so that it was not carried out for the other loops. Its effect, we checked, would not have altered the status of the other difficulties significantly.

If no other difficulties of a more serious nature had been present, we should have gone to the trouble of applying this correction. But precisely such a difficulty was encountered in the fact that the slope was not minus one. Various simple modifications of the theory were tried for allowing a slope $\neq -1$ but so far none has been found.

3.4. SECOND-ORDER THEORY

The difficulties listed above are only the major ones. They all seemed to point in the same direction, namely, that at the intensities used—and it would not have been practical to use lower intensities and correspondingly longer exposure times—the main approximation used in the theory was not valid. This approximation stated that for low intensities the incoming quanta were sufficiently separated in time so that the photoelectrons of one quantum would have a reasonable chance of being "alive" when the next photoelectron was generated, but a negligible chance when the third one came.

Efforts were made to extend the theory to include not only the interaction of successive quanta but also of the next ones. Apart from the difficulty of this attempt, the main discouragement came from the fact that such a generalization would introduce a large number of new parameters. With enough parameters, however, one can always fit the experimental results and there is little guarantee that the results of such a fit are meaningful. Therefore a further development of the theory in this direction was abandoned.

4. Conclusions

The research described in the second part of this report led to the following conclusions.

I. The theory on the basis of which the work on sequence loops was undertaken has a number of quantitative shortcomings which are a direct consequence of the nature of the problem, and no practical way is presently seen to remedy them.

II. The theory seems to be qualitatively useful in determining the order of magnitude of the quantities it set out to determine, namely, E_H and α .

III. The determinations lead to values of E_H , the number of quanta or photoelectrons required for a developable speck, in the range of 3-20 under the conditions investigated. The values of E_H depend on the density, the grain size, the type of emulsion, and the development conditions, within the range of the experimental variables investigated.

IV. Frequently (although not without exception), a trend seems observable for the less sensitive grains (in terms of quanta incident on the grain) to be more efficient in regard to the building of the developable speck, i.e., higher E_H' values correspond to lower E_H values and vice versa.

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