THE GRADATION AND INERTIA OF HURTER AND DRIFFIELD : A STUDY IN THEORY

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Summary

In a deep emulsion the shape of the D–log E characteristic depends on two quantities, the photon number P, which is determined by the number of photons required to form a development centre, and the ratio K of the absorption cross sections of a silver halide crystal before and after development. The position of the curve on the exposure axis is determined by the mean reaction cross section s of the crystals. In a coating a fourth quantity enters, the " opacity before development " of Hurter and Driffield, here called the exposure ratio R of front to back surface.

If development changes K and only K, the Hurter and Driffield laws governing gradation and inertia automatically follow. Experiments have shown that P, s, and R are independent of development in one or two emulsions, but the extension of this to all emulsions depends on the general validity of the two laws which Hurter and Driffield themselves established.

In a Schumann plate the ratio D/D_s, where D_s is the saturation density, remains constant as development proceeds and the two laws are still valid.

I. TWO EMPIRICAL LAWS

May 31, 1960 is the seventieth anniversary of Hurter and Driffield's paper (1890), describing their *Photochemical Investigations on Photographic Plates*, a paper which still ranks as the most important ever written on the photographic process.

As the first scientists to investigate the photographic plate, Hurter and Driffield had to establish the experimental facts before they could attempt a theory. They always used white light and this handicapped them, for many important laws, which have emerged in recent years, and particularly the Kaiser transform (1948), are valid only in monochromatic light. The two laws, however, which are valid in white as well as monochromatic light, Hurter and Driffield discovered.

(a) Gradation of Density

When a plate is given two different exposures, and these produce densities D_1 and D_2, the " gradation " or density ratio D_1/D_2 is independent of the time of development, and of the developer too, provided the latter is " well balanced ". And in the sense here used, all commercial developers are " well balanced ", for Hurter and Driffield meant chiefly that the developer shall not contain excess bromide, or any other substance which will dissolve some of the silver halide (Ferguson 1920, p. 95 ; hereafter quoted as MV).

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In support of this law Hurter and Driffield gave some measurements made on Ilford and Wratten plates (Table 1). They showed too that the law is valid after intensification (MV, p. 99), but not after reduction (MV, p. 100).

TABLE 1

SOME MEASUREMENTS BY HURTER AND DRIFFIELD (MV, p. 96) WHICH ILLUSTRATE THEIR GRADATION LAW

| Plate | Exposure M.C.S. | Development | | D_1/D_2 |
		4 min	12 min	
Ilford Red Label ..	10	0·77	1·26	1·63
	20	1·00	1·66	1·66
	40	1·18	1·96	1·66
	80	1·25	2·08	1·60
Wratten Drop Shutter..	10	1·17	1·74	1·50
	20	1·67	2·37	1·42
	40	2·06	2·91	1·41
	80	2·26	3·33	1·47

(b) The Inertia

When the so-called straight part of the characteristic is produced to cut the exposure axis, the point of intersection is written $\log i$, and i is called the " inertia " of the plate. Experiment shows that when either the time of development or the composition of the developer is changed, the inertia remains the same. Thus a " Manchester Slow " plate was developed first in eikonogen and then in hydroquinone (Fig. 1), and the time of development was altered in each experiment, yet the inertia was practically the same in all four determinations.

II. HURTER'S THEORY

Hurter devoted much time to theory, but he failed to explain the two laws, and no later writer has been more successful. Yet the general line the argument should take is clear from a careful reading of Hurter's own work. For Hurter (MV, p. 108) showed that the exposure E_v reaching the virgin crystals is a fraction $\exp(-Na_c)$ of the exposure E entering the surface of the emulsion,

$$E_v = E \exp(-Na_c). \tag{1}$$

Here N is the number of crystals already fertilized, or in Hurter's words (MV, p. 108) " the number of particles already changed ", while a_c is the absorption cross section of a halide crystal and the small volume of gelatine immediately surrounding it.

By implication Hurter also well understood that in the developed plate his " transparency ", a quantity identical with the " transmittance " of B.S. 1636 (British Standards Institution 1950), is

$$T = e^{-D} = \exp(-Na_g), \tag{2}$$

K

where the first equation defines the natural density **D**. True, the relationship between density and the absorption cross section of a silver grain

$$\mathbf{D} = Na_g \tag{3}$$

is generally attributed to Nutting (1913), who derived it from theory, and to Trivelli (1946), who verified it experimentally. True too, that the equation is not known to occur in any of Hurter's papers. But equation (1) does occur, expresses the same principle, and is much more recondite. Moreover, Hurter
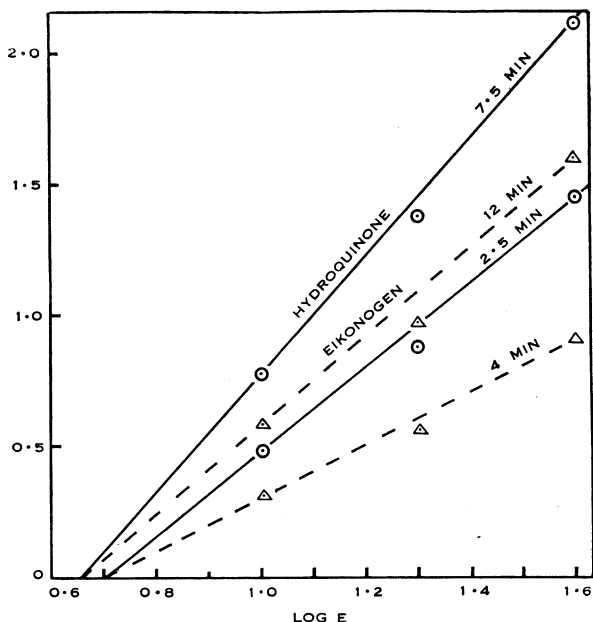

Fig. 1.—A Manchester Slow plate was given three different exposures and cut into four pieces. Two were developed in hydroquinone, one for $2\frac{1}{2}$ and the other for $7\frac{1}{2}$ min, and two in eikonogen for 4 and 12 min respectively. The densities are " exclusive of fog, which was very considerable ". In this figure Hurter and Driffield chose chords rather than tangents (MV, pp. 97, 120).

habitually used " weight of silver " and " density " as interchangable terms, even to the point where his editor felt bound to enter a caveat (MV, pp. 99, 100), so the reader can scarcely escape the conclusion that the Nutting equation was known to him.

On combining the above equations, the exposure reaching the virgin crystals is seen to be

$$E_v = Ee^{-K\mathbf{D}}, \tag{4}$$

where K is the ratio of the absorption cross section of a halide crystal before and after development.

$$K = a_c/a_g. \tag{5}$$

This is possibly a more suggestive and significant way of stating Hurter's result than any he himself employed, but it can scarcely be said to advance his account of absorption one iota.

Yet these equations are very close to a solution of the problem Hurter was investigating. For during development K decreases as a_g increases; and if of the parameters which govern the form of the characteristic, all but K are determined at the instant of exposure and are unaffected by development, the density may be written

$$K\mathbf{D}=f(E), \tag{6}$$

where $f(E)$ is invariant during development. Given this equation, the laws of gradation and inertia are easily shown to follow.

Seeing how near Hurter himself came to a solution, the further history of the subject is curious. By 1920 (MV, p. 34) experiment had shown that Hurter's equation of the characteristic is erroneous, but analysis had failed to locate the error; and this has remained true to the present day (Mees 1942, 1954).

After 1920 two lines of work can be distinguished. The Rochester school (Silberstein 1922, 1923; Silberstein and Trivelli 1930, 1938, 1945; Webb 1939, 1941, 1948) based their work on Silberstein's light dart hypothesis, and regarded as axiomatic the Poisson equation, which had already been shown to explain the characteristic produced by α-particles. True, Silberstein (1928) showed that the characteristic of a monolayer cannot be explained unless either the quantum theory or the Poisson equation is rejected. Although this conclusion was quite correct (Candler 1960), it was more than Silberstein himself was able to accept, and for another 20 years both quantum theory and Poisson law continued in general use.

Over the same period astronomers (Sampson 1923; Baker 1925, 1926, 1928; Wilson 1953) and spectrochemists (Kaiser 1941, 1948; Hughes and Murphy 1949; Morello 1955; Candler 1956; Arrak 1957) sought a transform linear in $\log E$. Generally, spectrochemists have been little interested in theory, but when Kaiser (1948) found a transform linear at all wavelengths, an exposure equation (Candler 1956) was immediately available, and analysis showed that the four parameters (Candler 1959) are precisely those selected by Hurter 70 years earlier.

Hurter's choice of the right parameters was a remarkable feat and reveals his intuitive grasp of a complex physical problem. For one might well expect with the Rochester school that grain size will alter the shape of the characteristic either directly or through its effect on the photon number.

In modern symbols Hurter's differential equation may be written

$$K\delta\mathbf{D}=(\mathrm{e}^{-K\mathbf{D}}-R^{-1})\delta(sE). \tag{7}$$

This states that the number of fresh crystals fertilized by a small additional exposure δE is measured by the increase in density $\delta\mathbf{D}$, and is proportional to the fraction $\mathrm{e}^{-K\mathbf{D}}$ of the additional exposure, which penetrates to the virgin crystals, less the fraction $1/R$ lost through the back surface, R being " the opacity of the unexposed plate " (MV, p. 110) or the exposure ratio of front to back

surface. The sensitivity of the crystals is determined by their reaction cross section s, which is in effect the reciprocal of Hurter's ε " the energy required to change one particle of silver halide into the condition capable of development " (MV, p. 109).

Whether the energy lost through the back surface should be deducted is doubtful, but no decision is necessary for an emulsion so deep that no light is lost through the back surface is an easier problem than a coating. And, once the equation of a deep emulsion is known, the equation of a coating is easily derived (Candler 1959). So the term $1/R$ may be dropped.
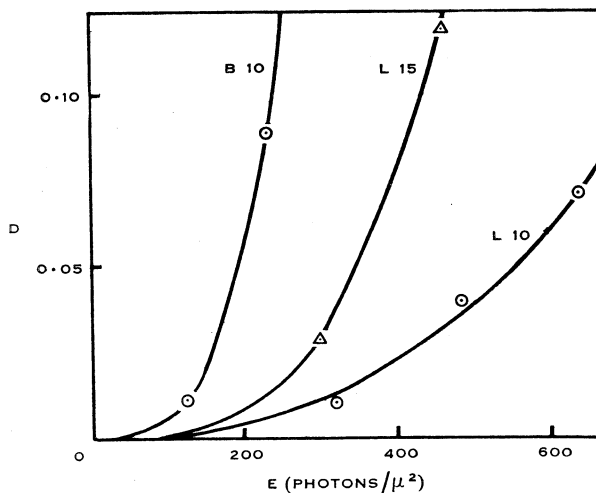

Fig. 2.—The density-exposure curves of three Kodak plates are all parabolic near the origin. These plates were exposed at 430 mμ and measured at Harrow in 1956.

For the rest, Hurter well understood that he had to know how the number of crystals fertilized varies with the exposure when the density is so low that absorption plays no part. Unfortunately, an unhappy extrapolation from high densities and an erroneous experiment (MV, p. 107) made him think that the number of crystals fertilized is proportional to the exposure, whereas later experiments (Fig. 2) have shown that the law is

$$D = kE^P, \tag{8}$$

where P is called the photon number, because it is most simply interpreted as the number of photons required to form a development centre. In most emulsions P lies between $1 \cdot 5$ and $3 \cdot 0$.

As Hurter believed P unity, he omitted it from his equation. When this error is corrected, his equation becomes

$$K \delta \mathbf{D} = e^{-PK\mathbf{D}}(sE)^{P-1}\delta(sE), \tag{9}$$

and on integration the exposure equation emerges

$$(sE)^P = e^{PK\mathbf{D}} - 1. \tag{10}$$

At low densities this equation cannot be distinguished from the author's equation (Candler 1959) (Fig. 3), which may be written either as

$$(sE)^P = PK\mathbf{D}e^{PKD} \qquad\qquad (11)$$

or

$$A + \log E = PKD + \log D, \qquad\qquad (12)$$

where $\mathbf{D}$ is the natural density $\ln(1/T)$ and D the common density $\log(1/T)$. At higher densities, however, differences occur, and a critical comparison is firmly in favour of the Hurter form. Nevertheless, Hurter's equation does possess one disadvantage; it is much more difficult to fit to an experimental curve. As either equation will explain the two empirical laws, the author's form will be used for the rest of this paper.
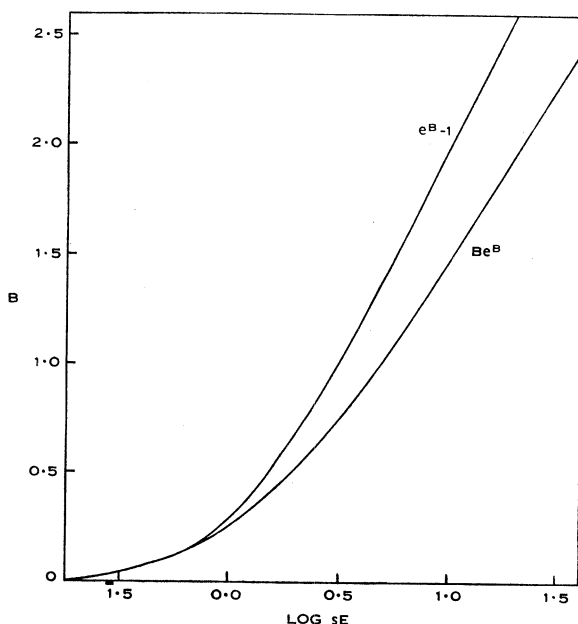

Fig. 3.—The Hurter characteristic of a deep emulsion is compared with one proposed by the author. B is an abbreviation for the product PKD. Both curves assume $P = 2$.

III. Invariance of Three Parameters

If theory is to be trusted, three parameters, the reaction cross sections, the photon number P, and the exposure ratio R of front to back surface, are all determined at the instant the development centres are formed, and only the fourth, the ratio K of the absorption cross sections, varies with development. This should remain true so long as the developer is one which neither aborts centres formed by the radiation nor develops grains which have not been fertilized by light.

Incidentally, this is a crucial test of theory. For, if the equation is really empirical and the theory illusory, all four parameters would surely vary.

Only one set of plates is known to have been developed for different times after being given the same exposure (Mees 1942, p. 409). The illuminant was white light, so one would not expect a monochromatic equation to apply. But Arrak (1957) noticed that a monochromatic equation is valid, presumably because the emulsion was panchromatic; and with white light the effective wavelengths are limited to a narrow range where K happens to be constant.
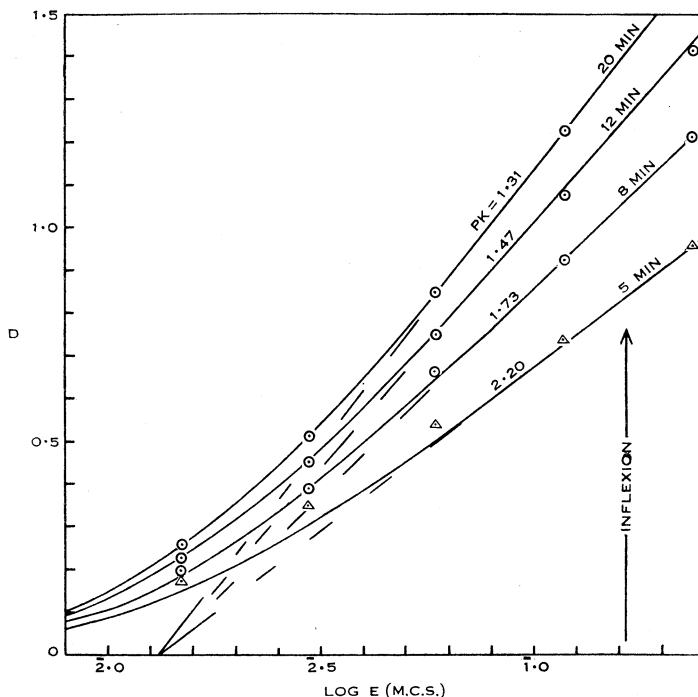

Fig. 4.—The characteristics of a panchromatic emulsion studied by Sheppard and Mees. The curves which follow theory, assume $P=2\cdot15$ and $\log(E_{MCS}/sE_\varphi)=\bar{2}\cdot07$; where the two exposures are in metre-candle-seconds and photons/μ^2 respectively. The circles are experimental. The development time and the value of PK are written beside each line.

The four characteristics can be fitted (Fig. 4) to an exposure equation with $P=2\cdot15$, and this confirms the invariance of P, which the author had noticed in a set of four B 10 plates developed, three in Ilford ID-2 of concentrations 33, 67, and 100%, and one in neat D.19b. In Sheppard's emulsion three curves of best fit have the same values of $A+\log PK$ (Table 2).

The development times were 8, 12, and 20 min, and a comparison of equations (11) and (12) shows that this establishes the invariance of the reaction cross section.

Hurter and Driffield's experiments were very extensive, and are still the substantial evidence that neither P nor s changes on development. In practice

small changes in s do occur in many developers, because the developer is not perfectly balanced, so some development centres are aborted or some grains not fertilized by light develop.

TABLE 2

THE PARAMETERS A AND PK FOR THE CURVES OF BEST FIT IN THE PANCHROMATIC EMULSION STUDIED BY SHEPPARD AND MEES

The photon number is taken to be $2\cdot15$

Development Time (min)	PK	A	$A+\log PK$
5	$2\cdot20$	$3\cdot68$	$4\cdot02$
8	$1\cdot73$	$3\cdot56$	$3\cdot80$
12	$1\cdot47$	$3\cdot62$	$3\cdot79$
20	$1\cdot31$	$3\cdot66$	$3\cdot78$

IV. THE GENERALIZED DENSITY

The shape of the characteristic depends on both P and K, and this makes the discussion of any general property laborious; but, when the natural density $\mathbf{D}$ is replaced by the natural form of the generalized density $\mathbf{B}$ defined as

$$\mathbf{B}=PK\mathbf{D}, \tag{13}$$

the shape of the generalized characteristic, or B–$\log E$ curve, depends only on P, and discussion is simplified. Inserting this quantity in the exposure equation,

$$(s\mathbf{E})^P=\mathbf{B}e^{\mathbf{B}}. \tag{14}$$

Convenient as natural logarithms and natural densities are in theoretical work, common logarithms and common densities are preferred for computation. Fortunately, the values of P and K are unchanged, and the above equation may be written

$$P\log(s\mathbf{E})=B+\log(B/M), \tag{15}$$

where $M=0\cdot434$. Here s remains the reaction cross section provided $\mathbf{E}$ is measured in photons per unit area; but if E is measured in arbitrary units s becomes a mere constant without physical significance.

Interest is largely concentrated on the B–$\log E$ characteristic, whose slope (Fig. 5) is

$$\mathbf{B}'=\mathrm{d}\mathbf{B}/\mathrm{d}(\ln E)=P\mathbf{B}/(1+\mathbf{B}), \tag{16}$$

or

$$B'=\mathrm{d}B/\mathrm{d}(\log E)=PB/M+B), \tag{17}$$

and whose curvature (Fig. 6) is

$$\mathbf{B}''=\mathrm{d}^2\mathbf{B}/\mathrm{d}(\ln E)^2=P^2\mathbf{B}/(1+\mathbf{B})^3, \tag{18}$$

or

$$B''=\mathrm{d}^2B/\mathrm{d}(\log E)^2=MP^2B/(M+B)^3. \tag{19}$$

Incidentally, one may easily show that

$$B=M\mathbf{B},\quad B'=\mathbf{B}',\quad B''=\mathbf{B}''/M. \tag{20}$$

As B, B', and B'' are all functions of sE, the generalized slope and curvature can be plotted for any assumed value of P.

In a coating the shapes of the characteristic and its two derivatives depend on the photon number and the exposure ratio of front to back surface, two quantities that are independent of the development time and the composition
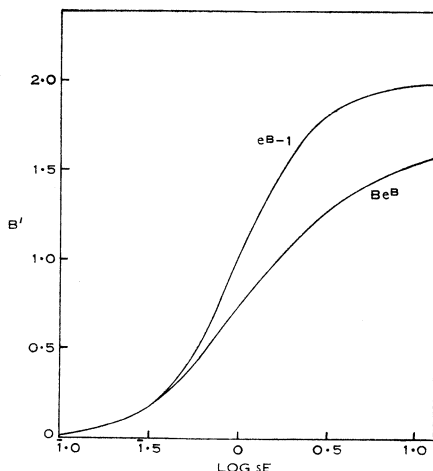

Fig. 5

Fig. 6

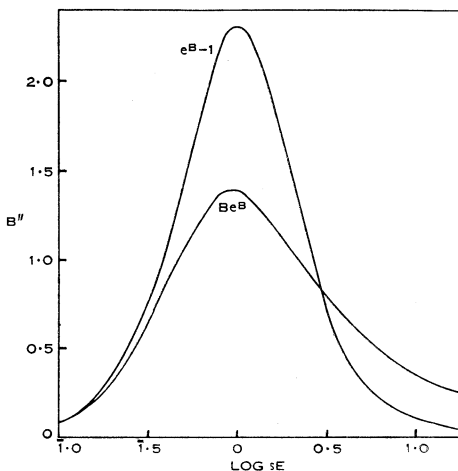Fig. 5.—The slope of the Hurter characteristic compared with an alternative proposed by the author.

Fig. 6.—The curvatures of the two characteristics similarly compared.

of the developer. Thus in a coating the generalized density may be expressed as a difference between front and back surfaces,

$$B_{ct}=B_f-B_b \tag{21}$$

This equation remains true when differentiated, so the slope and curvature of a coating may also be expressed as differences :

$$B'_{ct}=B'_f-B'_b, \tag{22}$$

$$B''_{ct}=B''_f-B''_b. \tag{23}$$

V. The Gradation of Density

When two plates are given the same exposure, the generalized density after development is the same even though the development is different. Accordingly, if the two plates are developed for times t_1 and t_2 and the resulting densities are D_1 and D_2,

$$B=PK_1D_1=PK_2D_2. \tag{24}$$

Hurter and Driffield's gradation law follows at once,

$$D_1/D_2=K_2/K_1, \tag{25}$$

for when the exposure is changed K_2/K_1 remains the same. Theory shows that in monochromatic light the gradation law is valid at all wavelengths, and this suggests a new method of verification. For if the effective exposures $s'E'$ at λ'

and $s''E''$ at λ'' are equal, the generalized densities will also be equal, since P is invariant in wavelength (Candler 1959),

$$D'/D''=K''/K'. \tag{26}$$

The size of the silver grains formed on development is independent of the wave length, so a_g' and a_g'' are equal, and

$$K''/K'=a_c''/a_c'. \tag{27}$$

In this form the gradation law can be verified experimentally only when the exposure is measured in absolute units, for only so can the effective exposures
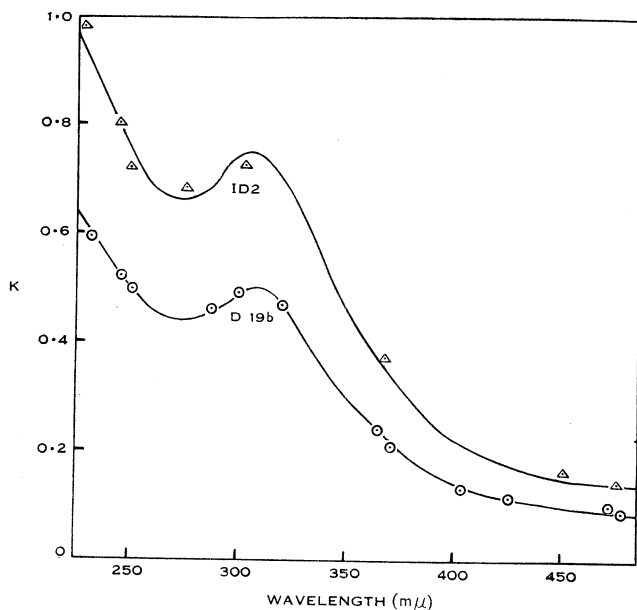

Fig. 7.—The variation of K with wavelength in two B 10 plates developed for 3 min, one in 33% ID-2 and the other in neat D.19b. In the empirical curves the density ratio varied from $1\cdot3$ to $1\cdot6$, but here the two curves have been adjusted until the ratio is $1\cdot5$ throughout.

$s'E'$ and $s''E''$ be made equal. But, if a change in the time of development or the composition of the developer changes the value of K at one wavelength from K_1' to K_2', and at another wavelength from K_1'' to K_2'',

$$K_1'/K_2'=K_1''/K_2''. \tag{28}$$

So the curves of K against wavelength should have ordinates in the same ratio when either the time of development or the composition of the developer is changed. This rule has been verified for a Kodak B 10 plate developed in Kodak D.19b and Ilford ID-2 (Fig. 7).

The gradation law remains valid after intensification (MV, p. 99) but not after reduction (MV, p. 100), and it is not valid if the developer contains excess bromide or any other agent which dissolves silver bromide. These observations

are simply explained if the law is valid only when the number of development centres is unchanged during development.   As the number of centres cannot be increased without producing fog or decreased without loss of speed, almost all commercial developers satisfy this condition.   The only important exception is the fine grain developer, which does in fact entail some sacrifice of speed.

VI. The Inertia

If the " straight " part of the characteristic is extended to cut the exposure axis at $\log i$, i is called the " inertia ".   The straight part may be written

$$D = \gamma \log E/i, \tag{29}$$

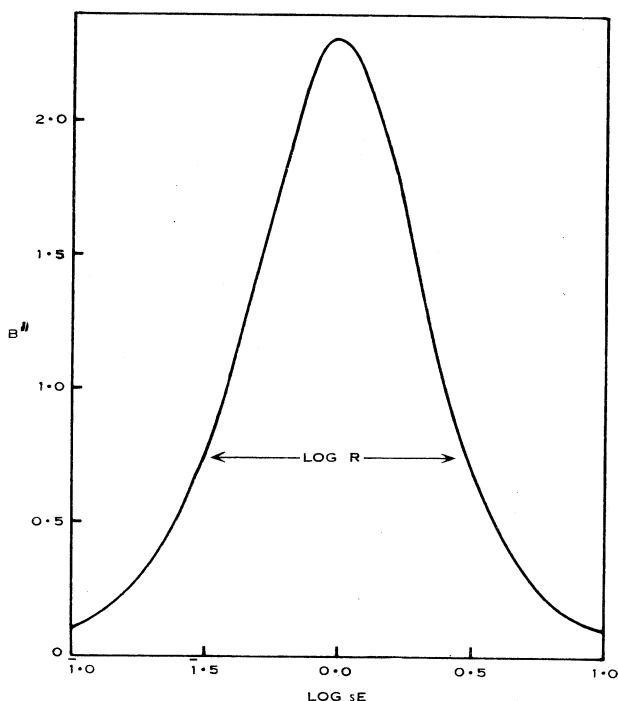where γ is the maximum slope.

Fig. 8.—At the inflexion of the characteristic of the coating the curvatures introduced by the front and back surfaces must be equal and be a distance $\log R$ apart on the $\log E$ axis.   Clearly there is only one exposure at the front surface which will satisfy both conditions.

When Hurter's theory (MV, p. 107) and Driffield's experiment (MV, p. 112) showed that no part of the characteristic is truly straight, Hurter identified the so-called straight part as the tangent at the point of inflexion X, or, in his own words, at " the point of double flexure " (MV, pp. 36, 222).   In the notation of the present paper, the equation of this tangent is

$$D - {}_xD_{ct} = {}_xD'_{ct} \log E/xE, \tag{30}$$

where $_xD_{ct}$, $\log {}_xE$ are the coordinates of the point of inflexion, and $_xD'_{ct}$ is the slope, commonly written γ. Accordingly, the intercept on the exposure axis is

$$\log i = \log {}_xE - {}_xD'_{ct}/{}_xD_{ct}. \tag{31}$$

This equation shows that the point of confluence lies on the exposure axis only if the exposure $_xE$ at inflexion and the ratio of slope to ordinate $_xD'_{ct}/{}_xD_{ct}$ are both independent of the time of development. These two conditions were recognized as necessary by Mees (1942, p. 409).

At inflexion the curvature of the coating is zero, so the curvatures due to the front and back surfaces are equal; and an examination of the B''–$\log E$ curve (Fig. 8) shows that for a given exposure ratio of front to back surface, this condition admits of only one solution. So at inflexion the exposure $_xE$ at the front surface, the generalized density of the coating, and its derivative $_xB'_{ct}$ are all uniquely determined. But by definition the generalized density is PKD, so at inflexion

$$_xB'_{ct}/{}_xB_{ct} = {}_xD'_{ct}/{}_xD_{ct}. \tag{32}$$

Thus the two conditions, which Mees recognized as necessary and sufficient, derive directly from theory.

While the inertia is precisely determined by the parameters s, P, and R, it cannot easily be made explicit in these quantities. In particular, the inertia is not simply related to the reaction cross section, which is the best measure of the speed of an emulsion, although not of a coating. Theory therefore gives no grounds for accepting the inertia as a measure of the speed, and this confirms an opinion long since reached empirically.

VII. WHITE LIGHT ILLUMINATION

Hurter and Driffield verified the invariance of gradation and inertia with white light. The theoretical proof given above applies only to monochromatic light.

Logically the natural line is from monochromatic to dichromatic illumination, and then by generalizing the results to white light; but so far as is known, not a single dichromatic characteristic is on record with which to check any calculations.

A preliminary study suggests that no difficulty occurs in passing from monochromatic to dichromatic illumination of a monolayer, provided the intensities of both radiations are measured in absolute units. From monolayer to deep emulsion should require no more than integration in depth, a procedure surely possible by numerical methods, even if the integrals are intractable.

A direct attack on the dichromatic characteristic of a deep emulsion is difficult, because the two radiations penetrate to different depths. If this does not completely bar the road, it is certain that the best line of attack is still obscure.

VIII. THE SCHUMANN PLATE

The characteristic of a Schumann Plate (Baker 1928; Candler 1960) has the same shape as that of a monolayer,

$$(sE)^P = D/(D_s - D), \tag{33}$$

where D_s is the saturation density. (Incidentally, this equation also describes the characteristics of five emulsions exposed to X-rays of wavelengths $0 \cdot 23$–$0 \cdot 63$ Å by Tellez-Plasencia (1954).)

As development continues one may expect the ratio D/D_s to remain constant along with s and P, so that the gradation law will be satisfied.

The slope of the Schumann characteristic is

$$\mathrm{d}D/\mathrm{d}\,(\log E) = PD(D_s - D)/MD_s, \tag{34}$$

where $M = 0 \cdot 434$. This is a maximum when $D = \frac{1}{2}D_s$, the slope being

$$\mathrm{d}D/\mathrm{d}\,(\log E) = PD_s/4M. \tag{35}$$

Thus the slope and ordinate at the point of inflexion increase together as development proceeds, and the inertia is constant, just as in a deep emulsion.

Note added in Proof

A dozen equations purporting to describe the characteristic have been published, but not one has claimed to describe the monolayer as well. In this Hurter's equation stands alone, for on differentiating (5) with respect to the depth, the equation of the monolayer (Candler 1960) emerges.

These two equations, monolayer and deep emulsion, describe the characteristic not only in the visible, ultraviolet, and X-ray regions of the spectrum, but also the characteristic of fast electrons, and the grain density along the track of an ionized particle. The evidence will be published shortly in the *Zeitschrift für wissenschaftliche Photographie*.

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