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DETECTION OF DEVELOPER BY-PRODUCTS USING THE SCHLIEREN METHOD

by

Stephen Kelly and Ralph Simon

An Abstract

A single-pass schlieren system, utilizing two 14 inch focal length lenses, has been developed for the purpose of demonstrating the feasibility of detecting and recording developer by-product flows from various photographic film/paper-developer combinations. The test samples studied were: Kodak Polycontrast F Paper in Kodak Developer, D-72 and Kodak Kodalith Contact Film in Kodak Developers D-11, Kodalith Super and Kodalith. The apparent amount of detectable flow varied greatly from test sample to test sample, but did not seem to show any significant shift in the pattern due to variations in the exposure conditions on the samples. Differences in the time of initiation of the flows were observed for the various samples. Selected photographs are included.

TABLE OF CONTENTS

LIST OF FIGURES

ACKNOWLEDGEMENT

I. INTRODUCTION

The purpose of the work described within this report is to demonstrate the feasibility of using a schlieren system to observe developer by-product flows produced during the processing of sensitized photographic materials. These by-products generally have the effect of retarding development to a significant degree in the area in which they occur.

In a case when two adjacent areas of an emulsion are given exposures of different magnitudes, the density levels in the immediate vicinity of the border may not reflect the actual effect of the radiation incident upon the emulsion. The result is generally known as adjacency effects. One of the most common effects is shown in the figure below, which is a microdensitometer trace across a boundry of a knife edge image. The increased density at the edge of the dense area is referred to as the "border effect" and the decrease in density in the low density area is referred to as the "fringe effect".
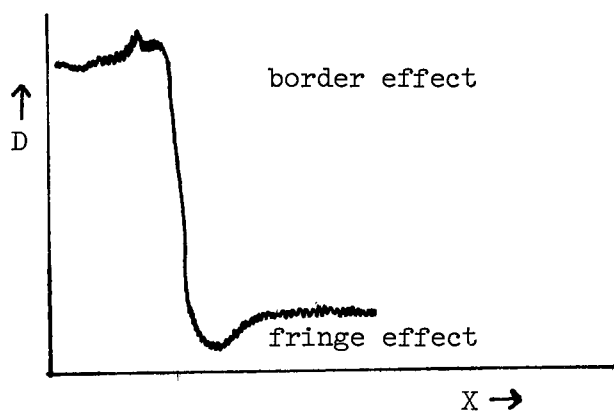
A. Mackie [1] noticed the latter effect in 1885, when a print was made of such an image described above. The resulting light lines on the print have been termed "Mackie Lines". The "border" and "fringe" effect taken together have been referred to as the "edge effect".

C. E. Ives and E. W. Jensen [2] believed that the above effect was due primarily too a local accumulation of partially or totally exhaused developer either in the emulsion or on the surface. They felt that this was a result of depletion of the developing agent and the resultant production of bromide by-product and they also observed that the non-uniformity was reduced or eliminated by vigorous random agitation. R. N. Wolfe and R. S. Barrow [3] felt that the effect was due primarily to the bromide formed as a by-product during development and not to the depletion of the developing agent.

There are some other related retardation effects which will be just mentioned here. The "Eberhard Effect" which becomes prominent when images of fine lines are produced. Decreased density in the region between two small images are known as the "Kostinsky Effect". A related effect is known as "bromide streaks" which are regions of decreased density caused by by-product streaming from images of high density. These by-products include bromides in high concentration. "Developer streaks" occur when comparatively fresh developer flows from low density images and hence increases the density in those areas over which the fresh developer flows.

In many commercial film-developer systems there are developers which contain bromides, but only one source of the by-product, the photographic emulsion during processing. Since this process is thought to be an internal as well as an external action, only the surface phenomenon are observable by optical methods. The simplest optical system for viewing such phenomenon would appear to be a schlieren system. The use of this system is based upon the premise that the developer by-products will have an index of refraction differing from the surrounding fresh developer and hence be detectable by a schlieren system.

II. The Schlieren Method

A. Theoretical Background

The schlieren method [4] depends upon the refraction of a light beam by gradiants in the refractive index of the medium through which the beam of light passes. In a typical system a straight edge, acting as a limiting diaphram, is so adjusted with respect to the edge of the light beam that refraction in one direction adds to the total illumination while refraction in the other direction subtracts from the total illumination. An image is formed which depends upon the differences of the refractive index in the light path.

The creation of a basic schlieren system entails critical optical alignment and careful selection of the system components. Figure 1 illustrates the components of a typical single-pass schlieren system. The purpose of the consenser lens is to image the source at the knife edge or slit. To insure uniform illumination of the image the source

should ideally have an extended ribbon filament and the condenser lens should be of high quality and well corrected. A collimated light beam is obtained by placing the first schlieren lens, f_1, a distance equal to its focal length away from the knife edge. The second schlieren lens, f_2, then focuses this collimated beam of light on to the plane of a movable knife edge, which controls the amount of illumination reaching the viewing plane. The test sample is placed between the two schlieren lenses, in the path of the collimated light beam. A disturbance or change in refractive index of the test sample will produce a deviation of the collimated beam and hence either add to or subtract from the illumination normally present on the viewing screen. In addition the image forming lens produces a real image of any object placed in the test sample area. The purpose of the knife edge is to control illumination on the screen and also the sensitivity of the system. This control is accomplished by shifting the knife edge so that in intercepts greater or lesser amounts of the light beam.

Two important factors of a schlieren system which must be considered are image contrast and system sensitivity. The image contrast, C, is defined as the change in illumination produced by the test sample divided by the original uniform illumination (without the test sample). This can be expressed as $C = \Delta I / I = f_2(\Delta E)/a$, where the following parameters are defined:

> $\underline{a}$ is the height of the slit image above the knife edge
>
> $\underline{E}$ is the angular deflection produced by the disturbance
>
> $\underline{f_2}$ is the focal length of the collimating lens.

If the image of the slit at the plane of the knife edge has a height b and the height of the slit image obscured by the knife edge is c, then the height of the slit image transmitted is equal to b-c=a. Therefore as the knife edge intercepts a greater amount of the slit image, the height of the image capable of transmitting the light beam decreases and thus the contrast increases. Sensitivity is defined as the change in contrast divided by the angular deflection produced by the disturbance or the ratio of f_2/a. Hence for a given ΔE and f_2 both contrast and sensitivity will be inversely proportional to the height of the slit image at the knife edge.

A brief mention is made here of the double-pass reflection schlieren system illustrated in figure 2. The main advantage of this system is that the light beam passes twice through the test sample, thus doubling the sensitivity. It does have one disadvantage and that is the reduction of resolving power due to the multiple path of the beam. The system illustrated in figure 2 was set up by Mr. Andrew Davidhazy (R.I.T. Distillation Research Lab.). The source is imaged by the condenser lens on the beam splitter which is located a distance equal to the focal length of the collimating lens away from the condenser lens. The test sample is located between the collimating lens and a plain front surfaced mirror which reflects the beam back to the reflecting surface of the prism. The beam is reflected 90 degrees and then imaged by a 35mm camera. The aperture of the camera lens serves as the second knife edge.

B. Experimental Procedure

The initial step consisted of building a basic schlieren system similar to the system illustrated in figure 1, using simple thin lenses for the optics. Evaluation of the basic aspects of the system was accomplished for two test samples; heat from a match and acetic acid dropped into water. A series of system modifications were made for each of the samples evaluated. These modifications consisted of:

1. lenses f_1 and f_2 were changed singly and in pairs
    from focal lengths of 28mm to 190mm.
2. replacing the first knife edge with a variable width slit
    capable of being aligned in either a vertical or horizontal
    position.
3. replacing both knife edges with variable diameter circular
    apertures.

The results of both tests confirmed the equations given earlier for sensitifity and contrast. The sensitivity of the system was increased when longer focal length lenses were used. The combination of the variable slit and knife edge provided greater sensitivity then either the combination of two knife edges or the two circular apertures. The sensitivity was also increased by decreasing the diameter of the circular apertures and decreasing the width of the variable width slit. One additional result of importance was the dependence of quality images upon optical alignment. Even with high quality optics, the alignment problem still remains the key to high quality images.

The above data was used to set up a system to evaluate the test sample described in section III. The optics consisted of a pair of thin 50mm lenses. These were later replaced with two relatively well corrected 50mm camera lenses. However, in neither case were any flow patterns observed coming from the test sample. Mr. Andrew Davidhazy

offered his services and set up the system shown in figure 2 and

discussed earlier in section IA. No flow patterns were observed

using this system either. Mr. Davidhazy then set up a second schlieren

system similar to the one illustrated in figure 1, but consisting of

the following components:

1. two achromat 60 inch lenses
2. the first knife edge was replaced by a circular aperture.
3. the second knife edge was replaced by the aperture of
   a camera lens
4. the image was formed by the camera objective on the
   film plane.

Results using this system were successful and are described on page

in more detail. However, it was rejected as a viable system due to

the long working distances required and the shortage of space in the

laboratory. The long distances required presented severe alignment

problems due to the fact that the components had to be mounted inde-

pendently of each other and critical alignment along the optical axis

could not be achieved. A solution could be effected if the components

could be mounted on a single optical bench.

This problem was alleviated by the aquisition of two achromat

14 inch lenses which could be mounted on a two meter optical bench.

A schlieren system was built which consisted of the following components:

1. lens f_1; Eastman Anastigmat Ektar 14", $f/6.3$
2. lens f_2; Wollensak Verifar 14", $f/6.0$
3. vertical slit of variable width and a vertical knife edge.

However, a nonuniformity problem became evident in the   image plane

when the knife edge was inserted in to the light beam. As the knife

edge traversed across the beam, severe nonuniformity was observed start-

ing on the side of the image field from which the knife edge was approach·

and continuing across the field.

This type of defect in illumination uniformity is essentially of the type expected according to the "Faucault Test". If a knife edge is passed slowly (right to left) through the image of the slit, the image field should darken uniformly and simultaneously if all the rays of the beam pass accurately through the focus. If the rays are not accurately focused, the field first becomes darkened over those regions from which rays pass to the right of focus. [6] In essence, if the optics are not capable of accurately focusing the vertical slit, it will be virtually impossible to obtain uniform illumination over the image field. Two options were immediately obvious as a solution to the focus problem. The first was the replacement of the source-condenser unit with a higher quality source and lens. No suitable replacement source was available, however, a jury rigged single filament source was tried with negative results. The original condenser lens was replaced by Rodenstock-Iioca-Ysarex 50mm, f/2.8 lens. This lens provided a slight improvement in the sharpness of the image of the slit and hence in the image field uniformity.

A third solution was replacement of the two 14 inch schlieren lenses, but this was impossible due to lack of adequate replacement lenses and insufficient time to properly mount such lenses if they were available. While setting up the system to begin photographing the flow pattern, it was noted that shifting the collimating lens to the right of its original distance, 14 inches, from the slit, improved the sharpness of the image of the slit. This resulted in a considerably improved image field uniformity, but as a cost of system sensitivity. An optimum compromise between slit image sharpness and image field uniformity was

achieved by shifting the lens 2.5 inches to the right. The aberrations responsible for this error were not immediately detectable. Figure 3 illustrates the final form of the schlieren system to be used. The camera components will be discussed in the following section.

C. Recording the Schlieren Image

If the schlieren field of test sample is to be recorded photo-graphically, then some consideration must be given to the optical relationships which fix the location and size of the image. For the purposes of this discussion the imaging system can be reguarded as consisting of the components illustrated below.
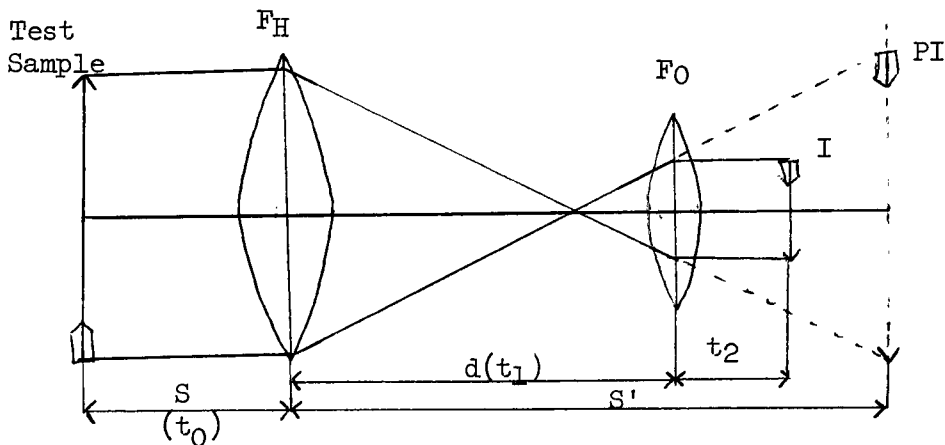
If the camera lens (F_0) were not present, the schlieren lens (F_H) would form a primary image (PI) of the sample at a distance S' from the lens. This distance can be computed from the "Gaussian" image equation, $\frac{1}{S} + \frac{1}{S'} = \frac{1}{f}$, to give a value of $S' = Sf/S-f$, where f is F_H in this case. If the camera lens is inserted into the system, the primary image then becomes the object of the camera lens and the above mentioned equation can be used to solve for the image or lens to film distance. This type of analysis is sometimes referred to the "successive imaging" analysis.

The total magnification for the system is simply the product of
the magnification of each lens considered separately, or $M_t=M_H x M_0$.
where $M_H=F_H/s-F_H$ and $M_0 = F_0/F_0+s'-d$. However, since $s'=sf/s-f$, s'
can be substituted in the above equation and when the multiplication
is carried out the total magnification becomes; $M_t = \dfrac{F_H F_0}{s(F_H+F_0)+d(F_H-s)-F_H F_0}$.

Alternatively the system can be considered from the point of view
of a ray trace method which consists of tracing a hypothetical ray
from the object to the image plane. This is accomplished by the use
of two basic equations applied in succession. First the transfer equation,
$Y_k=Y_{k-1}+(t_{k-1})U_{k-1}$; and second the refraction equation, $U_k=U_{k-1}-Y_k/F_k$. If
a ray is considered to start from the optical axis and returns to the
axis at the image, then the total magnification can be considered as
the ratio of the angle the ray makes with the first optical element to
the angle the ray makes with the last optical element or U_0/U_{k-1}. For
the system described above this ratio becomes U_0/U_2.

Calculations of the value of the magnification were made for both
methods (see Appendix B) with an average value of approximately 0.76.
These values were obtained with the following optical components in
the system:

> F_H is a Wollensak 14 inch, f/6.0 lens
> F_0 is a Spiratone 135mm, f/2.8 lens plus a
> Spiratone 2X Telextender (total focal length
> of 10.6 inches)
> s=t_0 = 15 inches
> d=t_1 = 15 inches
> Nikormat 35mm camera.

In addition to the size and location of the source, the spectral nature of the illumination had to be considered. For reasons to be discussed ahead, a Kodak Wratten Filter, Number 29 was used for source filtration. Since this filter transmits only from 6100A to well into the infrared region, it was decided that an extended red sensitive film would be best for recording the image. Normal panchromatic emulsions have sensitivities which extend usually to around 6500A. The film selected was Kodak 2475 Recording Film (Estar-AH Base) which is an extremely high speed panchromatic film with extended red sensitivity (out to about 7250A).

III. The Test Sample

The selection of our test sample was based upon two reasons; the first was the requirement for a blue sensitive emulsion to make it easier to handle and process while in the darkroom and the second was that it be a high contrast graphic arts film. Edge effects due to developer by-products play a great role in graphic arts processes due to the fact that the production of line, halftone and continuous tone images constitute a large part of many graphic arts processes. The film selected was Kodak Kodalith Contact Film (Estar Base), a high contrast, blue sensitive film on 0.004 inch Estar Base.

The test sample to be observed consisted of a strip of the Kodalith Film 1 by 3 cms. which was inserted into a spectrophotometer cell 1 by 1 by 4 cms. containing Kodak Kodalith Super Developer. A knife edge

exposure was placed on one half of the film along the long dimension. The sketch below illustrates the test sample placement. The idea was that observations could be made of the flow at the top of the apex of the film strip.

optical axis
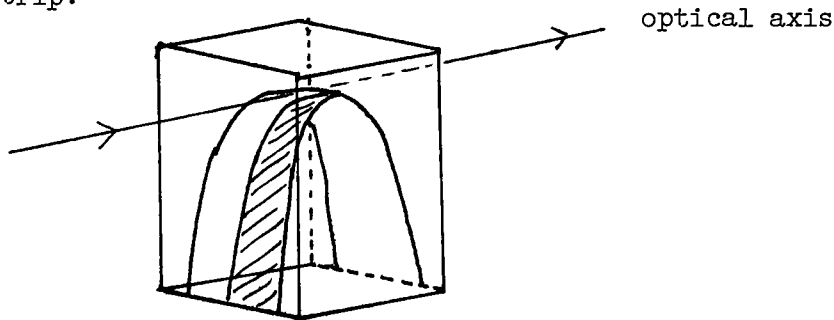
However, before any observations could be made, a preliminary problem appeared. This problem took the form of fogging of the film while in the presence of both the developer and the illumination. Despite the fact that the film was not sensitive to illumination trans- mitted by a Wratten 29 filter at "normal" levels of illumination, there was appearantly more than enough illumination striking the film to cause fogging. A test was run to determine the maximum illumination allowable before the film became fogged. The test consisted of simply placing an unexposed piece of film in the developer, while in the light path, for the maximum recommended development time of 4 minutes. Densitometry measure- ment on the samples indicated that a relative level of 1.4 footcandle of Wratten 29 illumination was the maximum allowable. This level was later found to be safe for several other emulsion-developer combinations also.

Additional problems soon appeared. Even if the image magnification had been great enough to enable the flow at the top to be seen, it is doubtful if any existed at the very top. The nature of the test sample orientation was such that appearantly the effect of gravity caused the

by-products to flow down the sides of the sample away from the apex which
was under observation. This hypothesis was advanced when several of out
test samples involving Kodalith Contact Film and super Kodalith Developer
showed wedge shaped edge effects along the boundry of the knife edge
exposure. The sketch below illustrates the effect observed.
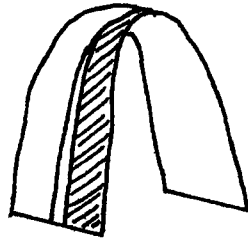
Similar results were obtained for a Kodak Kodabromide F-1 Paper
in Kodak Developer, D-72(1:2), hence the proposal of using a curved
piece of film or paper as described above seems to be unworkable due
to the above mentioned problem. Several tests were run using Kodabromide
F-1 Paper in D-72(1:2) with the system described on page 5. A 1x1 inch
piece of paper was placed in the developer (unagitated) in a vertical plane
and later in a horizontal plane. Two exposure conditions were evaluated
for each case; a total exposure and a knife edge exposure. A downward
flow pattern was observed for the totally exposed piece of paper both
in the vertical and horizontal position. No flow pattern was observed
from the paper having the knife edge exposures both in the vertical and
horizontal position. It would appear in the case of the knife edge ex-
posures that the unexposed areas of the paper had the capability of
absorbing the by-products produced in the exposed area and hence are
not able to diffuse out of the emulsion and into view. Since photographs
were not taken the sketch below illustrates what was observed. (The

system used to observe this flow is described on page 7.)
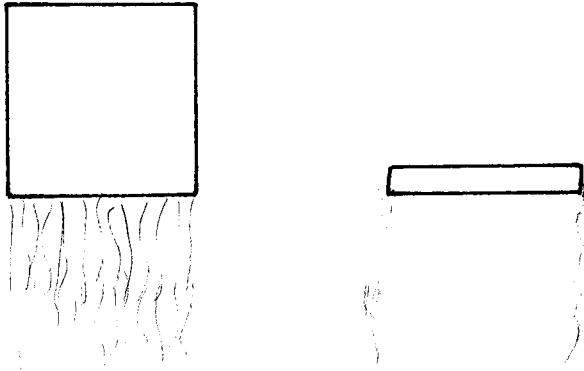
Flow Patterns Observed

The system actually established (see section IB and IC) was used

to observe the following test samples:

1. Kodalith Contact Film in Super Kodalith Developer
2. Kodalith Contact Film in Kodalith Developer
3. Kodalith Contact Film in Kodak D-11 Developer
4. Kodal Polycontrast F in Kodak D-72(1:2) Developer.

Three exposure conditions were evaluated for each test sample; no

exposure, total uniform exposure and a knife edge exposure on one half

of the sample. The sample size was one centimeter square. Photographs

were taken on Kodak 2475 Film using a 270mm lens on a Nikormat 35mm camera

at 1/250 second at f/2.8. Test sample 4 (above) was processed in Kodak

Developer, DK-50 for 8 minutes at 70 F. The other test samples were

processed in Kodak Developer, D-19 for 12 minutes at 70 F. This was

done to provide greater contrast in the negative images. All film was

processed using a Kodak Roll Film Tank with agitation for 5 seconds every

30 seconds.

Time did not permit a complete evaluation of interpretation of

the results, but a brief summary of the observed phenomena can be made.

The combination of Polycontrast Paper and D-72 Developer produced

by-product flow patterns that were by far the most distinct. The nature

and apparent quantity of the by-product flow did not appear to be signifi-

cantly different for each of the exposure conditions examined. One

observation that appears significant is that a flow was observed even

for the unexposed piece of paper. The time of initiation of the flow

did show a change with respect to exposure conditions, although not

enough data was available to draw any meaningful conclusions. The avail-

able data is summarized below.

Exposure Condition	Flow Initiation Time
Knife edge exposure:	
on right	5 seconds
on top	15 seconds
on bottom	30 seconds
Total exposure	15 seconds
No exposure	5 seconds

Photographs were taken at development times of 20, 30, and 40 seconds.

A series of prints are enclosed illustrating the development of the flow

pattern with development time. Prints 1 - 3 show the flow from a piece

of paper with a knife edge exposure on the top half of the paper for

development time 20, 30, and 40 seconds.

The test sample consisting of Kodalith Contact Film in D-11 Devel-

oper produced flow patterns that were about at the same level of de-

tectability as the previous sample. Once again the flow pattern appeared

the same for exposure conditions consisting of total exposure, no exposure

and a knife edge exposure on the right. However, the initiation of the

flow remained constant at 40 seconds for all the exposure conditions.

Prints 4 - 6 show the flow pattern as it appeared at 40 seconds for each

of the exposure conditions mentioned above.

The test samples consisting of Kodalith Contact Film in Kodalith Developer and Super Kodalith Developer were evaluated for only two exposure conditions, total and none. In the case of the Super Kodalith Developer the unexposed film did not show evidence of a flow until 1.5 minutes had elapsed. The pattern extended down well below the bottom edge of the film indicating that the flow had started prior to visual detection. In the case of the unexposed film the pattern was detectable after 40 seconds, but once again showed signs of having started prior to that time. For the latter the flow could not be detected directly from the camera viewfinder, but were visable only on the photographs.

The totally exposed film in Kodalith Developer produced flows which were directly visible and photographically recorded. No flow patterns were detectable for the unexposed film either visually or photographically. The flow patterns for both Kodalith developers were very faint in appearance.

IV.   Conclusions

A schlieren system utilizing two achromat 14 inch lenses was built for the purpose of detecting and recording developer by-products formed during the development of selected photographic emulsions. The system was successfully demonstrated for four film/paper-developer combinations. The schlieren system was not operated at optimum conditions due to an inadequate source and inferior quality lenses. A better source would be one that consists of single extended ribbon filament such as found in an arc lamp. Ideally custom designed lenses should be used for a schlieren system, but since this is usually impossible the only alter-

native is to use only the highest quality lenses available which are compatible with the individual system.

V. Recommendations

The first suggestion resulting from this report would be to build a more precise system based on a single extended ribbon filament and well corrected lenses. Optimizing these two factors will reduce, if not eliminate completely, the non-uniformity in the image field. Since this system was only a feasibility or demonstration study, future work could be done with the intent of identifying the by-products. This could possibly be done by correlating the angular deflection of the light beam by the medium with the index of refraction of the same medium. Relative concentrations of these by-products could possibly be obtained by making density measurements on a negative image of the flow pattern. All of these would probably entail some additional modification of the schlieren system described above.

VI   Footnotes

1   A. Mackie, "British Journal Of Photography", 64(1918),
      11.

2   C. E. Ives and E. W. Jensen, "Journal SMPTE", 40(1943), 107.

3   R. N. Wolfe and R. S. Barrows, "PSA Journal", 13(1947), 554.

4   Eastman Kodak Company, "Schlieren Photography", Phamplet
      Number P-11, 1964.   .

5   F. Twyman, "Prism and Lens Making", Jerell-Ash Co.,
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Appendixes

A. Numerical Aperture and Resolving Power

Image resolution can be considered as the ability to see clearly

two adjacent lines or points. The ability to do this is limited by

the diffraction pattern which results from having a finite aperture

in a system. If you consider a system which images two points in

object space, the image is in the form of Airy Disc. If the points

under consideration are close enough the two patterns will overlap and

it is this degree of overlap of patterns that determines resolution.

A usual criteria for the minimum separation of the two points is called

Rayleigh's Criterion for resolution and is taken as 0.61 /N.A., where N.A.

is called the numerical aperture of the optical system being used to

image the points and is equal to n sinθ. θ is the cone angle of either

the object or image and n is the index of the lens. The resolving power

is just the reciprocal of the minimum separable distance and is equal

to n sinθ/0.61 . Hence maximum resolving power is obtained with maximum

lens aperture. The Airy Disc pattern illustrating the above situation is

shown below.
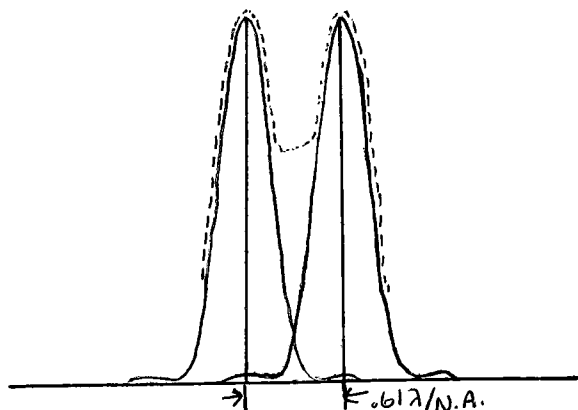
B. Calculations for Total Image Magnification

1. Successive Imaging Method

$$M_t = - \frac{F_H F_O}{S(F_H + F_O)+d(F_H-S)-F_H F_O}$$

Where: F_H = 14 inches
F_O = 10.6 inches
S = 15 inches
d = 15 inches

So: $M_t = - \frac{14(10.6)}{15(14+10.6)+15(14-15)-14(15)}$

$M_t = -\frac{148}{206} = - 0.72$

2. Ray Trace Method

$M_t = U_0/U_2$

Assume: $Y_0=0$, $U_0=1$.

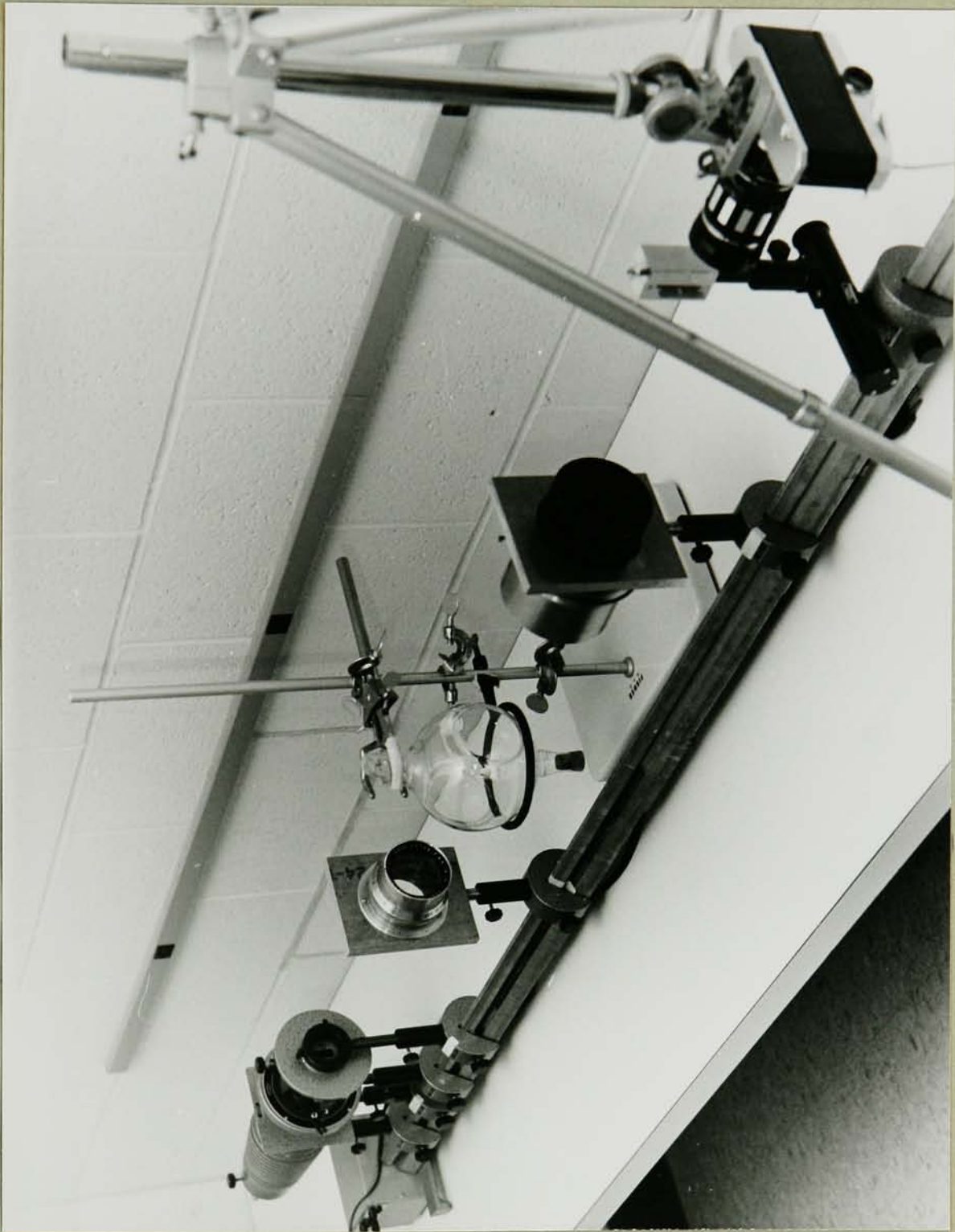Given: F_1 = 14 inches, F_2=10.6 inches, to = 15 inches,

t_1=15 inches.

then: $Y_1=Y_0+t_0 U_0=15$

$U_1=U_0-Y_1/F_1 = 1-15/14=-0.07$

$Y_2=Y_1+t_1 U_1 = 15+15(-0.07)=13.95$

$U_2=U_1-Y_2/F_2=-0.07-13.95/10.6=-1.39$

therefore, $M_t=U_0/U_2= 1/-1.39 =-0.79$

C.   PHOTOGRAPH OF THE SCHLIEREN SYSTEM

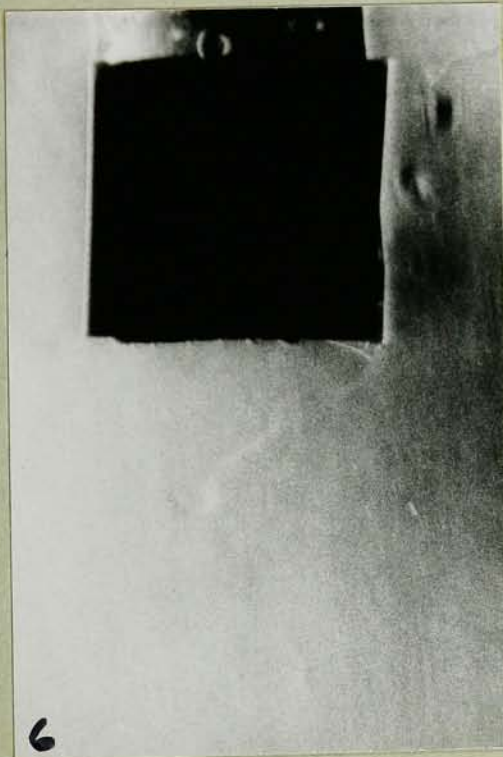D.   PRINTS OF SELECTED FLOW PATTERNS
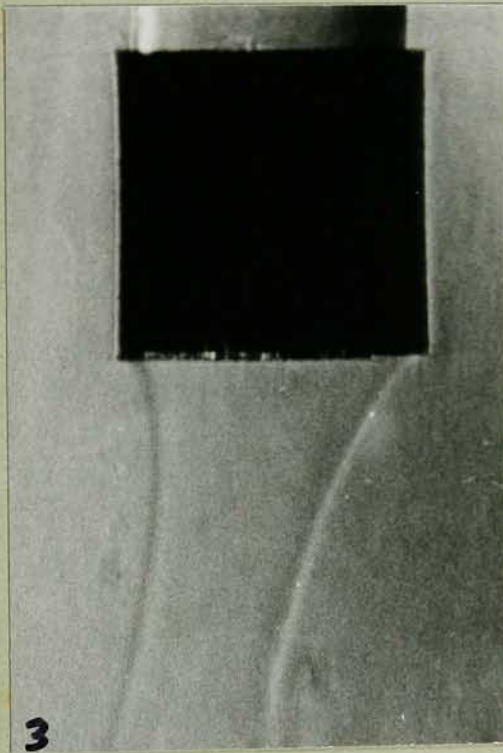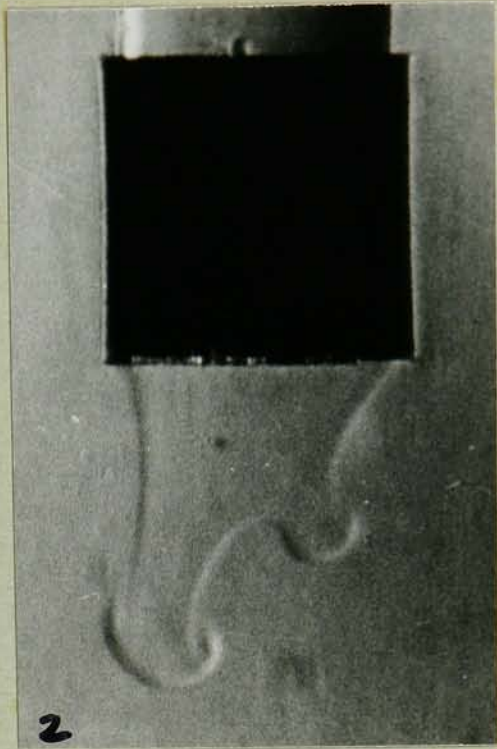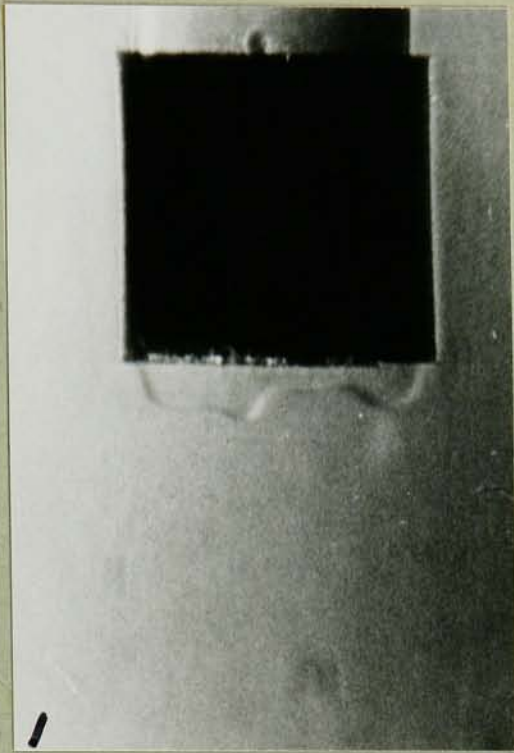
PRINTS 1, 2, 3

TEST SAMPLE: Kodak Polycon-
trast F Paper in Kodak
Developer D-72

Knife edge Exposure on
top half

PRINT 1:   20 seconds
            development time

PRINT 2:   30 seconds
            development time

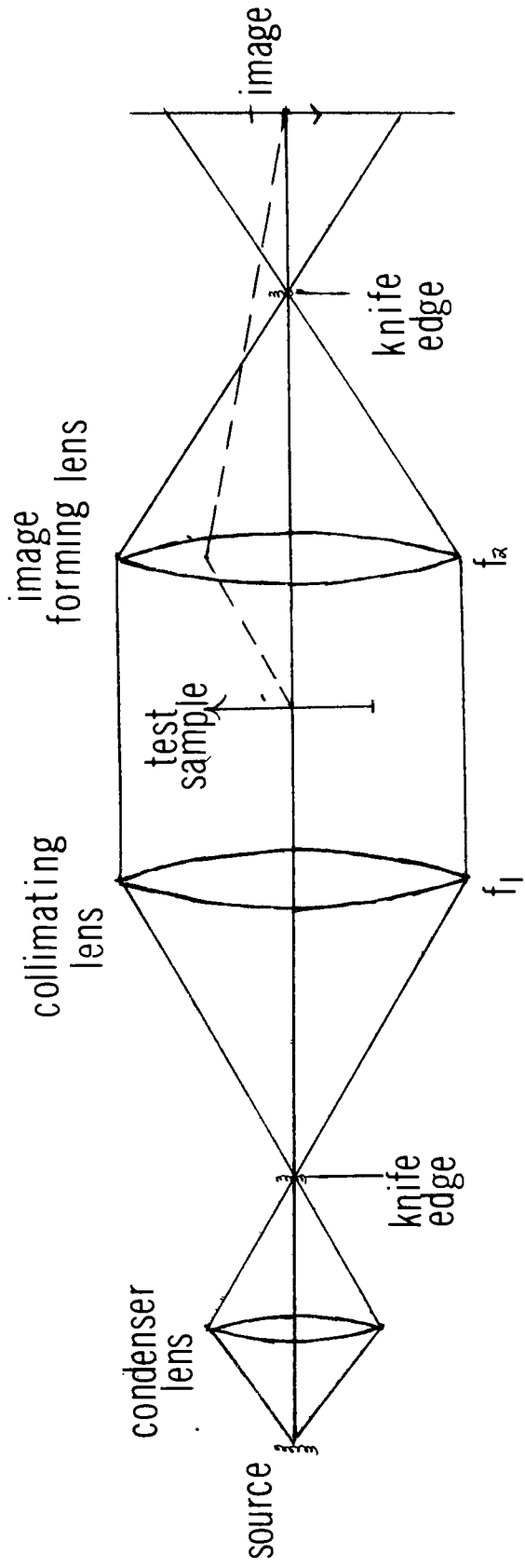PRINT 3:   40 seconds
            development time

4

5

6

PRINTS 4, 5, 6

TEST SAMPLE: Kodak
Kodalith Contact Film
in Kodak Developer D-11

DEVELOPMENT TIME: 40 seconds

PRINT 4: Total Uniform
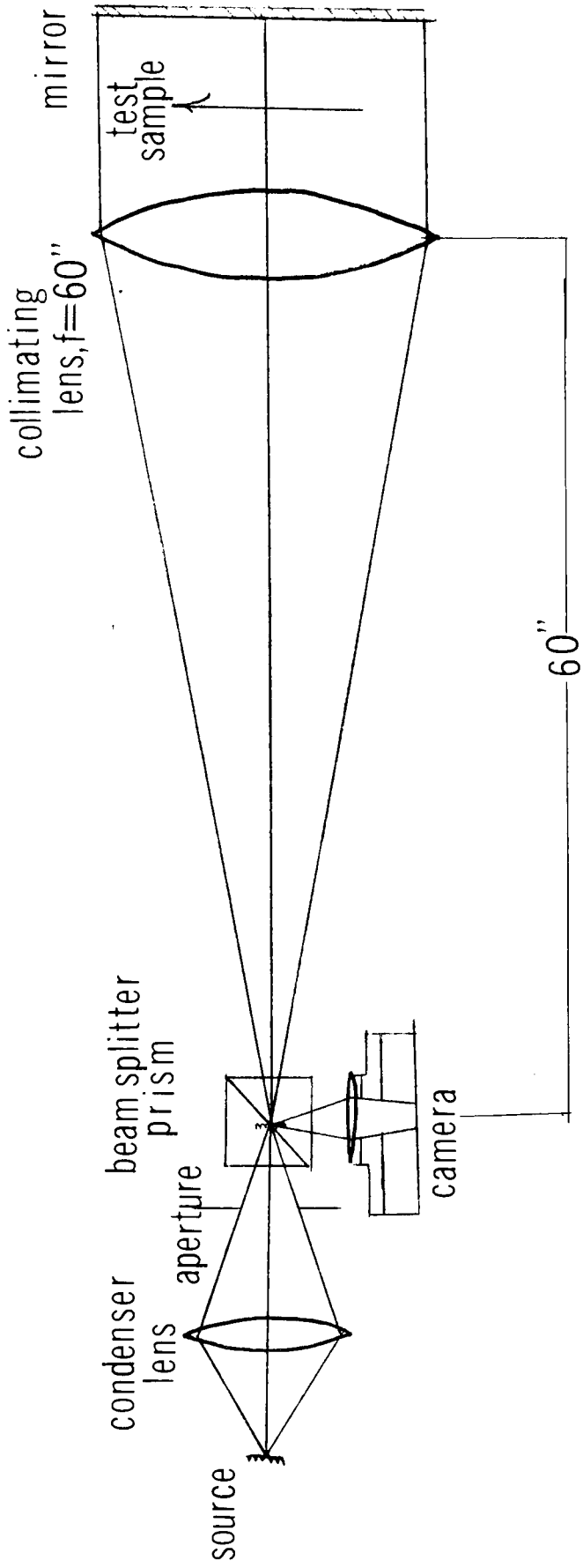   Exposure

PRINT 5: No Exposure

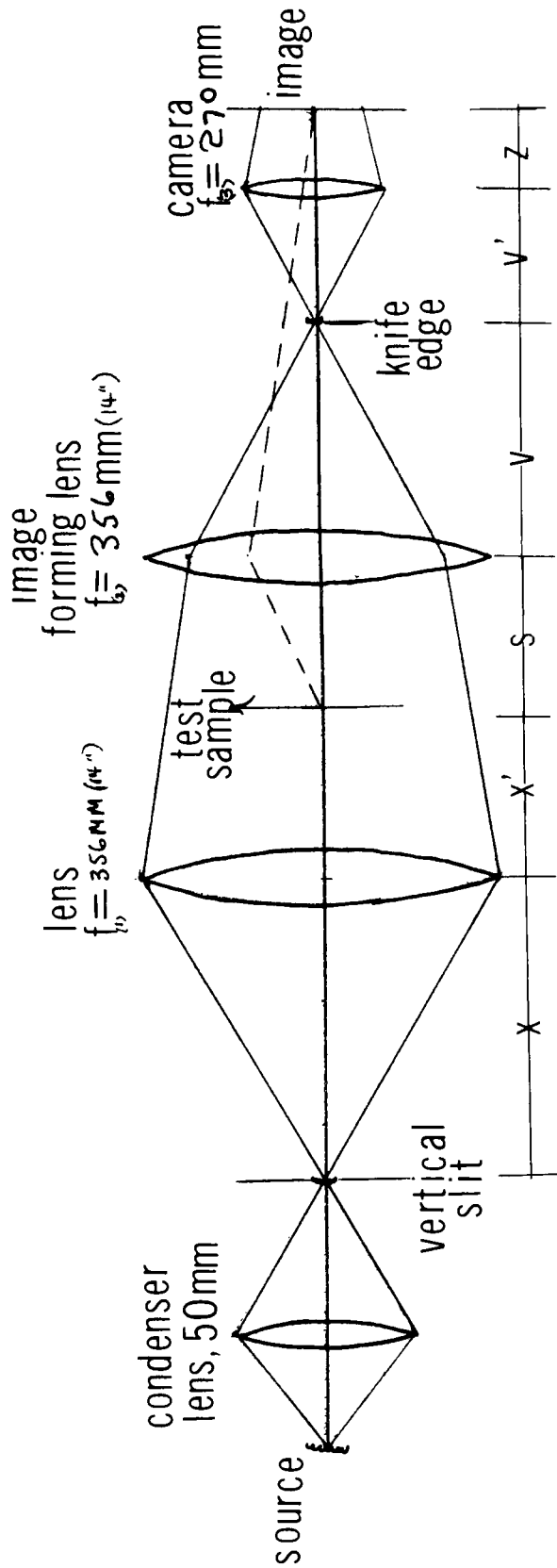PRINT 6: Knife edge
          exposure on right
          half

Basic Single-Pass Schlieren System

Figure 1

Basic Double-Pass Reflection Schlieren System

Figure 2

Single Pass Schlieren System Used

Figure 3