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## SHOCK-INDUCED LUMINESCENCE (U)

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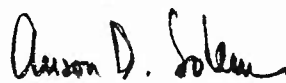


SHOCK-INDUCED LUMINESCENCE

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ABSTRACT: When a 270 kilobar shock wave emerges from an illuminated aluminum surface in a vacuum, blue light is emitted. This phenomenon is affected significantly by surface conditions. A chemically-formed aluminum oxide surface layer on the metal luminesces brightly for at least 0.64 microseconds. Normally oxidized aluminum surfaces luminesce less brightly for a much shorter time ( $\sim 0.01$  microseconds). The effect of non-oxidized material on the phenomenon is shown in that shocked gold does not luminesce. The luminescence from aluminum is attributed to electrons, dragged by the shock (acoustoelectric effect). The electrons reach the aluminum oxide layer and, falling into the ionized F' centers, emit blue light.

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The detonation of solid explosives affords a means of exerting high pressures unattainable by any other convenient means. This very rapid application of energy in the form of a shock wave has some very important effects on the structure and behavior of metals, semiconductors, and insulators. The basic understanding of these effects is important in many ordnance applications. The Explosions Research Department has undertaken a research program to study this important field. As a result luminescence has been detected from an aluminum surface on the emergence of a strong shock wave. The result was first reported by the authors at the American Physical Society Meeting in Detroit, Michigan, March 23, 1960. This report describes fully the experimental evidence and theoretical interpretation of this phenomenon. The work was done under Task FR 52, Dynamic Properties of Solids, in the Laboratory's Foundational Research Program. This report is for information only and should not be construed to represent the final opinion of this Laboratory.

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## SHOCK-INDUCED LUMINESCENCE

## INTRODUCTION

1. The response of solids to shocks and their associated pressure pulses is important in the understanding and use of such materials. Shock waves deform and change solids, i.e. compress them, (1) change their elastic constants, (2) and produce changes in phase. (3) (4) They must also change the electronic configuration of the solids and add or generate imperfections, such as vacancies or dislocations, in passage through the solids. A wide number of shock induced phenomena such as shock forming, (5) shock etching (6), shock welding (7), shock sintering, etc. have been observed and used. This report describes a set of experiments in which shock luminescence of an aluminum oxide surface has been observed. Shock luminescence is defined here as the emission of light from such a surface when an intense shock wave emerges onto the surface. This light is interpreted as the result of electrons, dragged along by the shock wave, falling into oxygen-ion vacancies in the aluminum oxide surface layer.

## 2. THE EXPERIMENTAL METHOD

## 2.1. The Phenomenon.

If a flat metallic plate is illuminated with a beam of light normal to the plate's surface, a certain intensity of light is returned from the plate, also in the normal direction, by reflection. Such light can be observed by a camera or other recording device located on the axis joining the source and the plate. The intensity of this light is constant under static conditions. In a dynamic experiment it has been shown, with the arrangement indicated above, that a discontinuous change occurs in the light coming from the plate's surface when a plane shock wave travelling through the plate emerges on the surface. (8) This might result from the shock producing a change in the surface geometry of the plate so that its reflection characteristics are changed, or it might be the result of some physical phenomenon occurring at the surface. For an aluminum plate an increase in light intensity occurs, and the present shock experiments were designed to determine if the increase is due to emission of light from the aluminum oxide surface layer.

## 2.2. The Test Specimens.

To study the shock luminescence of illuminated aluminum oxide surface layers, 61S aluminum plates 25-mm thick were used as test specimens. One surface of each plate was chemically polished with a solution of orthophosphoric acid, sulphuric acid, and chromic acid to form a thick irregular aluminum oxide surface layer. (9) On this face three cold-rolled aluminum foil strips, 0.01-mm thick, and one gold leaf strip, less than 0.01-mm thick, were arranged with a spacing as shown in Figure 1. This arrangement provided a heavily oxidized surface (the treated surface of the plate), normally oxidized surfaces (the aluminum foils), and a non-oxidized surface (the gold leaf).

## 2.3. The Test Arrangement.

The experimental arrangement for these tests is shown in Figure 2.

Detonation of the explosive system causes a shock wave which has a peak pressure on the order of 270 kilobars to enter the plate. This wave traverses the plate and on emerging at the plate's free surface is plane-parallel to the free surface to within  $\pm 0.35$  mm across the 50 mm central region of the specimen plate.

The free surface moves forward into a vacuum with a pressure on the order of 1 micron of mercury. (This vacuum eliminates the possibility of a luminous air shock developing at the rapidly moving plate surface.)\*

The free surface of the specimen plate traverses a void, 1.60-mm thick, and then impacts a fixed glass witness plate.

The specimen is illuminated with normally incident light from an argon flash bulb through the half-silvered mirror. The light is synchronized with the explosive system so that the surface is illuminated for about 10 microseconds before arrival of the shock.

The light coming from the surface of the plate in a direction normal to the surface is observed by the 70 mm rotating mirror smear camera after reflection at the

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\* Experiments done in air and helium show the same light intensity increase when the shock wave emerges from the free surface as the experiments done in a vacuum.

half-silvered mirror and passage through an optical filter, Wratten No. 44 (4600 to 5200 Å transmission band).\* The slit of the camera is arranged to view along a narrow line across a diameter of the specimen plate that covers both foils and treated surface of the plate. The camera records a time resolution on the order of 10 milli-microseconds of the light intensity for points on the slit line. Figure 3 shows a typical film record obtained in the experiments.

#### 2.4. Measurement of Reflectivity Coefficients.

The observations of these experiments are of light reflected from or emitted by the surfaces of the test specimens and how this light changes as shocks pass through the surfaces. These observations are made in terms of calibrated initial coefficients of reflectivity of the surfaces and measurements, from the photographic records, of the change of these coefficients during the experiment; a change of light recorded during the experiment being taken as a change of reflecting coefficient.

The initial reflectivity coefficients,  $R_0$ , of the surface of the specimen plate, the aluminum foils, and the gold leaf, are determined by a Lummer-Brodhum photometer.<sup>(10)</sup> The light entering the instrument is filtered by the same filter used in the dynamic experiment.

The dynamic changes in the reflectivity coefficients that occur during the experiments are obtained from densitometer readings of the smear camera film records. The measurement of these changes is made quantitative by the use of a calibrated, photographic step-wedge (Figure 3) attached to the film. This is done to correct for the possible failure of the reciprocity law when the film is exposed to short duration intense light. The step wedge is placed on the film so that it calibrates the film response before light changes occur when the shock wave emerges from the specimen free surface.

The analysis of the records and calibrations will be discussed later.

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\* Three auxiliary experiments were performed using color photography without the filter to determine the color of the argon light and the light emitted from the aluminum oxide surface.<sup>(11)</sup>

### 3. EXPERIMENTAL RESULTS

#### 3.1. Interpretation of the Records.

Eleven experiments have been carried out investigating the shock luminescence of aluminum surfaces, eight using Tri-X negative super speed film and three using Ektachrome color film. The positive print of a typical black and white record is shown in Figure 3. In the analysis the record is divided into two time regions by the arrival of the shock wave at the free surface of the specimen, the first abrupt changes in optical density of the film reading from left to right, Trace A. To the left time is designated negative, to the right time is positive.

In negative time the three bright streaks running horizontally across the record are light reflected from the aluminum foils while the dull streak is light reflected from the gold leaf.

The thickness of the foils is so small that the shock transit time through them is negligible as can be seen by examination of the shock wave arrival discontinuity.

Examination of the shock arrival discontinuity shows shock luminescence and the effect of surface preparation on this phenomenon:

The increase in the light intensity that occurs at the aluminum or aluminum oxide surface fails to appear at the gold leaf surface.

The increase in light intensity persists into positive time and the duration of persistence depends upon surface characteristics. A short period of high light intensity is noted for the aluminum foils; a long interval is seen at the chemically treated surface.

It is to be noted that the uniformity of the discontinuity in the transverse direction excludes jets, spalling of the foils from the plate, or spalling of the free surface of the plate as causes for the increase in light intensity.

The second abrupt change in light intensity, in positive time, records the impact of the specimen surface on the glass retaining plate. On the color films this change is signalled by a color change from blue to an intense yellow which is believed due to sodium emission caused by impact

shock heating of the sodium in the glass plate. The mean velocity of the free surface across the gap was 2.76 mm per microsecond indicating a shock pressure of 270 kilobars<sup>(1)</sup> in aluminum.

### 3.2. Analysis.

The quantitative results of the change in light intensity reaching the smear-camera film at zero time is given in terms of the effective reflectivity coefficient,  $R$ . The term effective is used to indicate that not only reflected but also emitted light is included, and that not only the electronic effect but also surface geometry and smoothness properties contribute to the change. In terms of measured data, the effective reflectivity coefficient is derived in Appendix A and given by

$$\log_{10} R = \log_{10} R_0 + f(\Delta d_m)$$

where  $R_0$  is the initial reflectivity coefficient and  $f(\Delta d_m)$  is the function determined by the film density change from negative to positive time and a calibration curve as shown in Figure 4. For the linear portion of this curve the equation above becomes

$$\log_{10} R = \log_{10} R_0 + 1.84 \Delta d_m.$$

For the record indicated in Figure 3 the values of the effective reflectivity coefficients are calculated for the middle section of the aluminum plate, for the aluminum foil below this section, and for the gold leaf. The middle section of the specimen was selected for analysis because the shock wave was plane in this region. Zero time was determined experimentally where the optical density first changed discontinuously. The optical densitometer had an aperture 0.1524 mm in diameter. Traverses were made with points 0.19 mm apart corresponding to 0.08 microseconds separation in time. Each point was read independently three times and the values were averaged arithmetically. The values of the effective reflection coefficients are plotted against time in Figure 5.

The chemically treated aluminum plate reflectivity coefficient shows an increase from 60.2 percent to 134.8 percent in a time interval of 0.15 microseconds. A reflectivity coefficient greater than unity requires that light be emitted from the surface.

The curve for the aluminum foil shows a rapid decrease at zero time. Examination of the smear camera record, Figure 3, shows a momentary increase in the light intensity, i.e., effective reflectivity coefficient, at zero time. The densitometer measurements did not resolve this increase. The ten other experiments also showed a momentary increase in light intensity at the aluminum foils on shock-wave arrival.

The gold leaf curve shows a very rapid decrease in the effective reflectivity coefficient when the shock reaches the free surface.

Examination of the curves in Figure 5 shows that the optical density of the film in negative time is constant. The light from the argon flash bomb is constant and no changes in the experimental arrangement occur during this period.

#### 4. THEORETICAL INTERPRETATION

Figures 3 and 5 show that the shock wave arriving at the surface of the aluminum plate produces luminescence. This phenomenon is interpreted in terms of the acoustoelectric effect (12), (13), (14) in metals and F' centers (15) (16) in the aluminum oxide surface layer. These two phenomena are explained briefly as follows:

The acoustoelectric effect is the transfer of energy from the mechanical shock wave to the free electrons in the metal. The shock wave represents energy which moves into the solid with shock velocity. Electrons moving slightly slower than the shock wave gain energy from this wave and are swept along with it. As a result a thin sheet of electrons move in synchronization with the shock wave. The shock generated in this experiment has a pressure of 270 kilobars and represents a power density of  $10^9$  watts per square cm.

The second solid state physics concept is the F' center in excess-metal oxides. In these oxides, oxygen ions,  $O^{2-}$ , are missing. Each vacancy represents two missing negative charges and two free electrons are trapped to compensate for this. An oxygen-ion vacancy with its two trapped electrons is called an F' center.

The shock-induced luminescence is explained as follows:

The blue light from the argon flash bomb illuminates the transparent aluminum oxide layer. A quantum of this light gives 2.64 electron-volts to an electron trapped in an F' center and the electron escapes, ionizing the F' center.

The excited electron wanders through the oxide layer and may be trapped in a low energy surface state from which it is emitted by the Auger effect<sup>(17)</sup>. Evidence<sup>(18)</sup> exists that the argon light removes the adsorbed layer of O<sup>-</sup> ions from the oxide surface and makes it easier for the electrons to leave the low-lying surface states.

After illumination by the argon light, an electrostatic gradient develops across the oxide layer. The probability is greater that the electrons near the surface leave the F' center traps and are emitted from the surface.

After the surface oxide layer has been excited by the argon light, the shock is generated by the explosive system. When the electron sheet dragged by the shock reaches the metal-oxide interface, the electrons spill over into the oxide layer by the tunnel effect, thermal emission, or electrostatic gradient. Some of these electrons fall into the singly ionized F' centers and emit quanta of 4700 Å (2.64 eV).

A qualitative check of the above mechanism has been obtained by studying the aluminum oxide layer and its effect on the luminescence. This irregular surface of the chemically treated aluminum plate luminesces brighter and longer than the smooth surface of the aluminum foils.

As a further check on the interpretation, it would be interesting to measure the emission of electrons from the aluminum oxide surface layer. Some of the free electrons in the oxide layer get trapped in the shallow surface states and are emitted. Electrons should leave the surface during the initial argon light illumination. A much more intense stream of electrons should leave the oxide surface when the shock-dragged electron sheet enters the oxide layer.

## 5. CONCLUSIONS

The experiments described here strongly indicate that internally illuminated aluminum oxide surfaces luminesce when subjected to 270 kilobar shock waves. This luminescence is a surface property of the material and can be optimized by forming an irregular, oxygen-deficient surface oxide layer by chemically treating the aluminum surface. The experiments prove that air shock and spalling do not explain the phenomena.

The luminescence shows a maximum in the blue portion of the spectrum, probably at 4700 Å. This luminescence is similar to the Kramer effect (18) and to the cold cathode emission effect (17). Therefore all oxygen-deficient oxides (i.e. zinc oxide, cadmium oxide, aluminum oxide, and gallium oxide) should exhibit shock luminescence. From the similarity to the other phenomena, there should be electron emission from the aluminum oxide surface during illumination by the argon light and during the shock-induced luminescence.

The yellow light emitted from the shocked glass is probably caused by sodium atoms excited by the shock wave. Investigation of this effect might lead to an estimate of the temperature of shocked materials.

## APPENDIX A

## EFFECTIVE REFLECTIVITY COEFFICIENT

The effective reflectivity coefficient  $R$  of a metal surface is determined in terms of experimentally measured data, i.e., the initial reflectivity coefficients of the surface and the optical density of the smear-camera record.

$R$  is defined as

$$R = I_r/I_i \quad (A1)$$

where  $I_r$  and  $I_i$  are the intensities of light reflected from and incident on the specimen surface. Multiplying numerator and denominator by  $I_{r0}$ , the light intensity before the shock wave arrives at the free surface, and taking logarithms one obtains

$$\log_{10} R = \log_{10} (\Delta R) + \log_{10} R_0 \quad (A2)$$

where

$$\Delta R = I_r/I_{r0} \quad (A3)$$

and

$$R = I_{r0}/I_i \quad (A4)$$

To determine the effective reflectivity coefficient,  $R_0$  and  $\Delta R$  must be known.  $R_0$  is measured by standard photometer methods. To evaluate  $\Delta R$ , one measures the change in optical density  $\Delta d_m$  of the smear-camera record from negative time to positive time. The optical density measurements are measured with an optical densitometer and this change is calibrated in terms of optical density measurements related to the optical step-wedge.

The optical density of an exposed and developed film is a unique function of the light energy incident on the film if the spectrum of the light and the development process remain invariant. These conditions are satisfied in each experiment. Furthermore the light energy is equal to the product of the light intensity times the interval of exposure. Since the smear camera exposes each part of the film for a constant interval, the relative density of the developed record is a function of light intensity only.

The change in optical density  $\Delta d_m$  is calibrated in terms of the optical step-wedge since the reciprocity relation fails. The optical density of the film under the optical wedge, say under step  $i$ ,  $d_{mwi}$ , is measured with the optical densitometer; so also is the optical density of the film in negative time which is not covered with the step wedge,  $d_{m-}$ . The change in optical density of the  $i$ th step,  $\Delta d_{mwi}$ , is given by the relation

$$\Delta d_{mwi} = d_{mwi} - d_{m-} \quad (A5)$$

This measured change is calibrated in terms of the optical density of the step-wedge. The light intensity transmitted by the  $i$ th step of the optical wedge,  $I_{cwi}$ , compared to the incident light intensity,  $I_o$ , is given by the density  $\Delta d_{cwi}$  where

$$\Delta d_{cwi} = \log_{10} I_{cwi}/I_o \quad (A6)$$

The values of  $\Delta d_{cwi}$  are correlated uniquely with  $\Delta d_{mwi}$  given analytically by

$$\Delta d_c = f(\Delta d_m) \quad (A7)$$

or plotted as in Figure 4.

The optical step-wedge method cannot calibrate measured densities greater than  $d_{m-}$ . Examining Figure 4 one sees that the calibration curve is linear for small changes below the  $d_{m-}$  value. Extrapolation of the linear curve into the region of positive density changes gives at worst the lower bound to the calibrated density change. The linear portion of Figure 4 satisfies the relation

$$\Delta d_c = 1.84 \Delta d_m \quad (A8)$$

$\Delta R$ , Equation (A2), is now evaluated in terms of measurable quantities. The calibrated change in optical density is

$$\Delta d_c = \log_{10} I_{a+}/I_{a-} = \log_{10} I_{d+}/I_{d-}, \quad (A9)$$

where  $I$  designates light intensity,  $+$  and  $-$  denote positive and negative time regions,  $a$  and  $d$  stand for densitometer measurements made after the experiment and events that took place during the experiment, respectively. Since the

experiment is designed to allow only the effective reflectivity coefficient to vary, one can write

$$I_{d+} = KR \quad (A10)$$

$$I_{d-} = KR_o \quad (A11)$$

where K is determined by the experimental arrangement (Figure 2) and is a function of the argon light intensity, the mirror, the glass plates, the optical filter, and the smear camera.

Substituting Equations (A10) and (A11) into Equation (A9) inserting (A3) and (A7) into the resulting equation and putting this into Equation (A2), one obtains

$$\log_{10} R = \log_{10} R_o + f(\Delta d_m). \quad (A12)$$

For the linear portion of the calibration curve given in Figure 4,  $f(\Delta d_m)$  is given by Equation (A8), and

$$\log_{10} R = \log_{10} R_o + 1.84 \Delta d_m. \quad (A13)$$

Equations (A12) and (A13) are expressions which give the effective reflectivity coefficient in terms of two measurable parameters: the initial reflectivity coefficient and the optical density of the smear-camera records.

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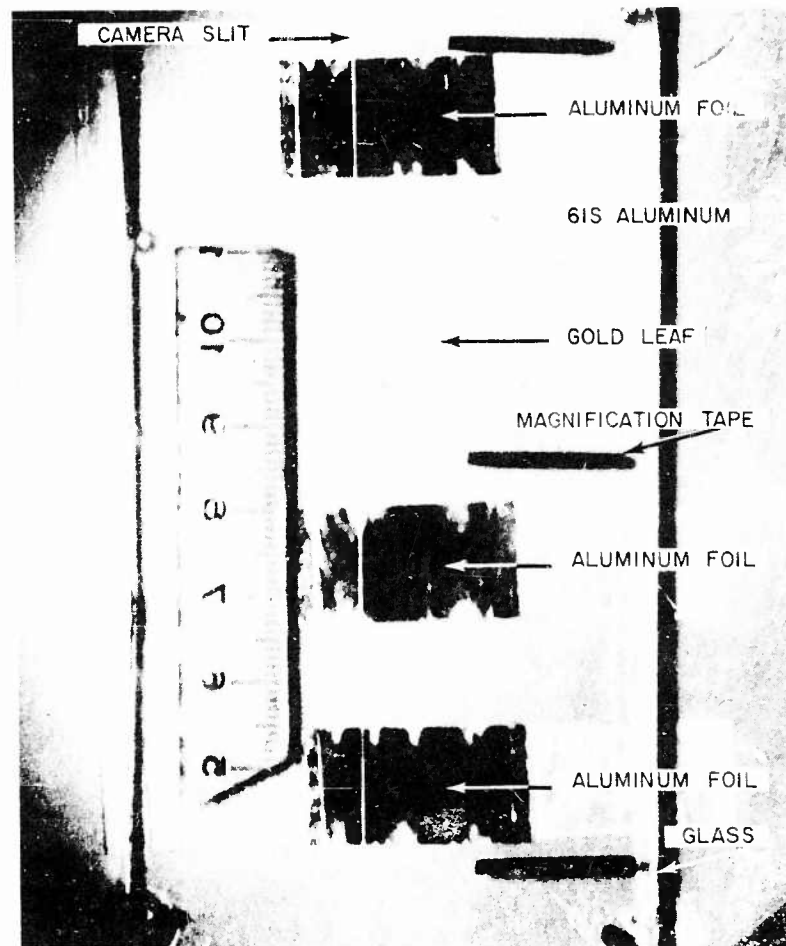


FIG. 1 SPECIMEN PLATE ARRANGEMENT

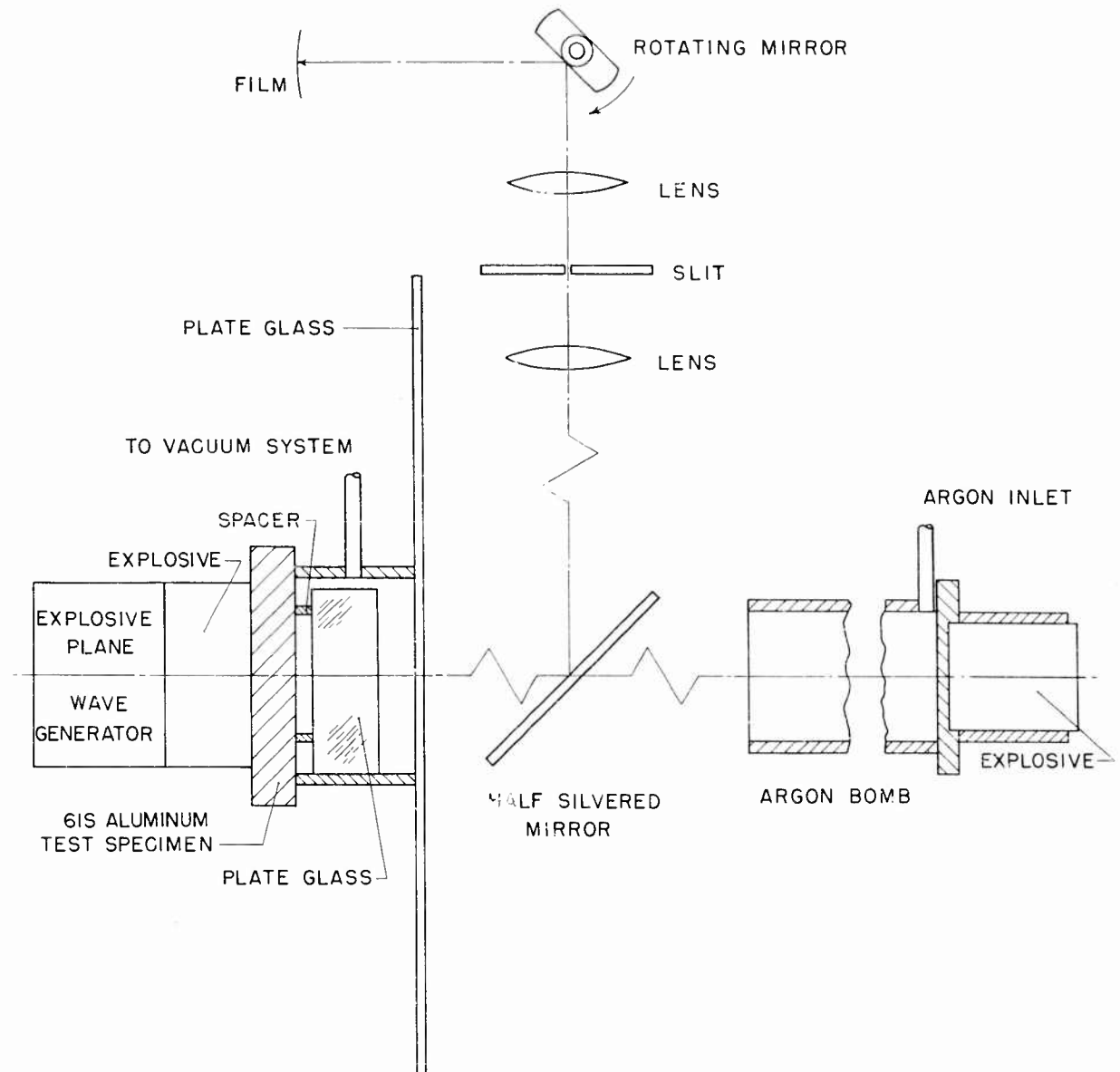


FIG. 2 EXPERIMENTAL ARRANGEMENT

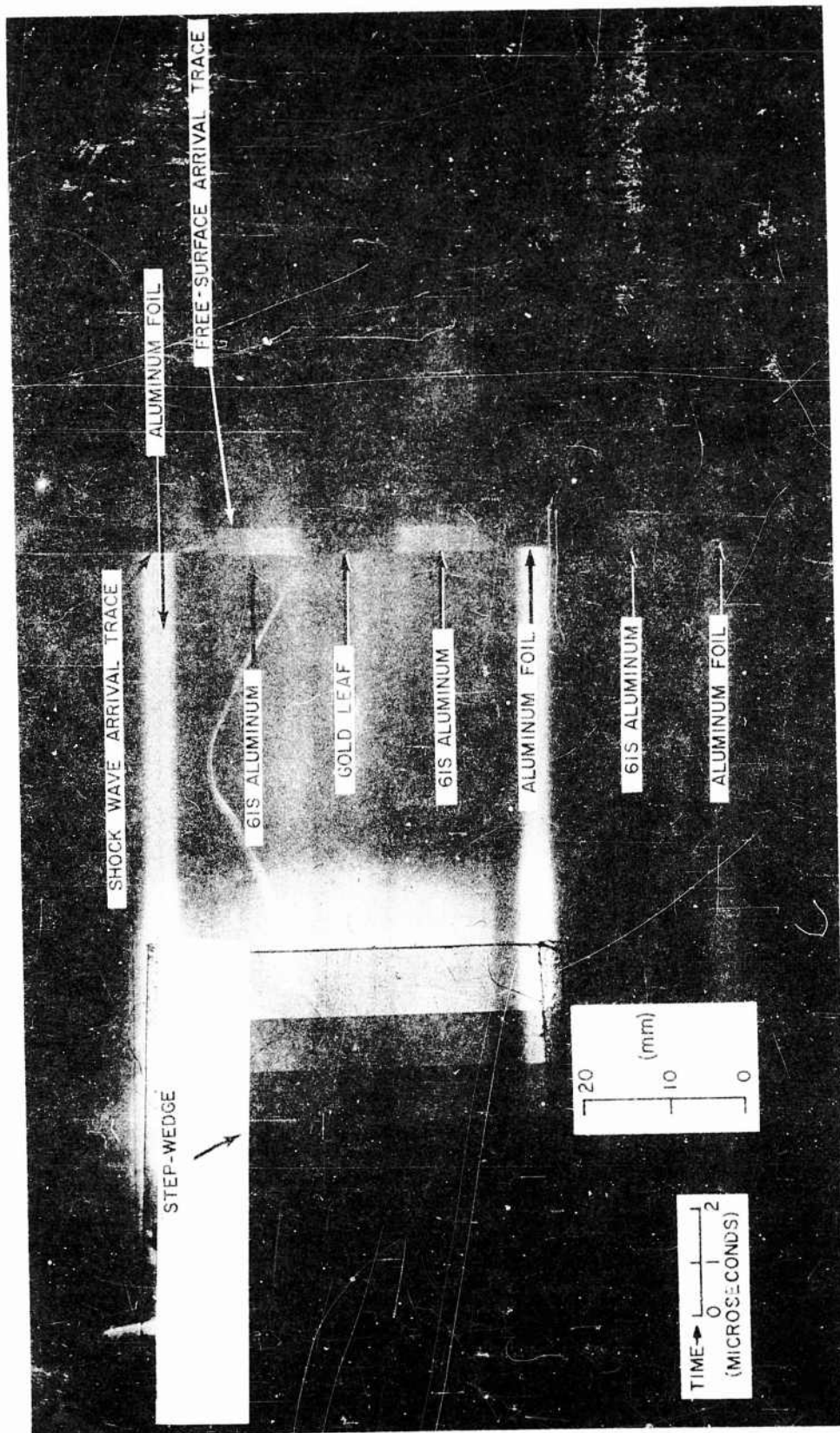


FIG. 3 SMEAR CAMERA RECORD CORRELATING INTENSITY CHANGES IN REFLECTED LIGHT WITH SHOCK WAVE ARRIVAL AT FREE SURFACE OF SPECIMEN PLATE.

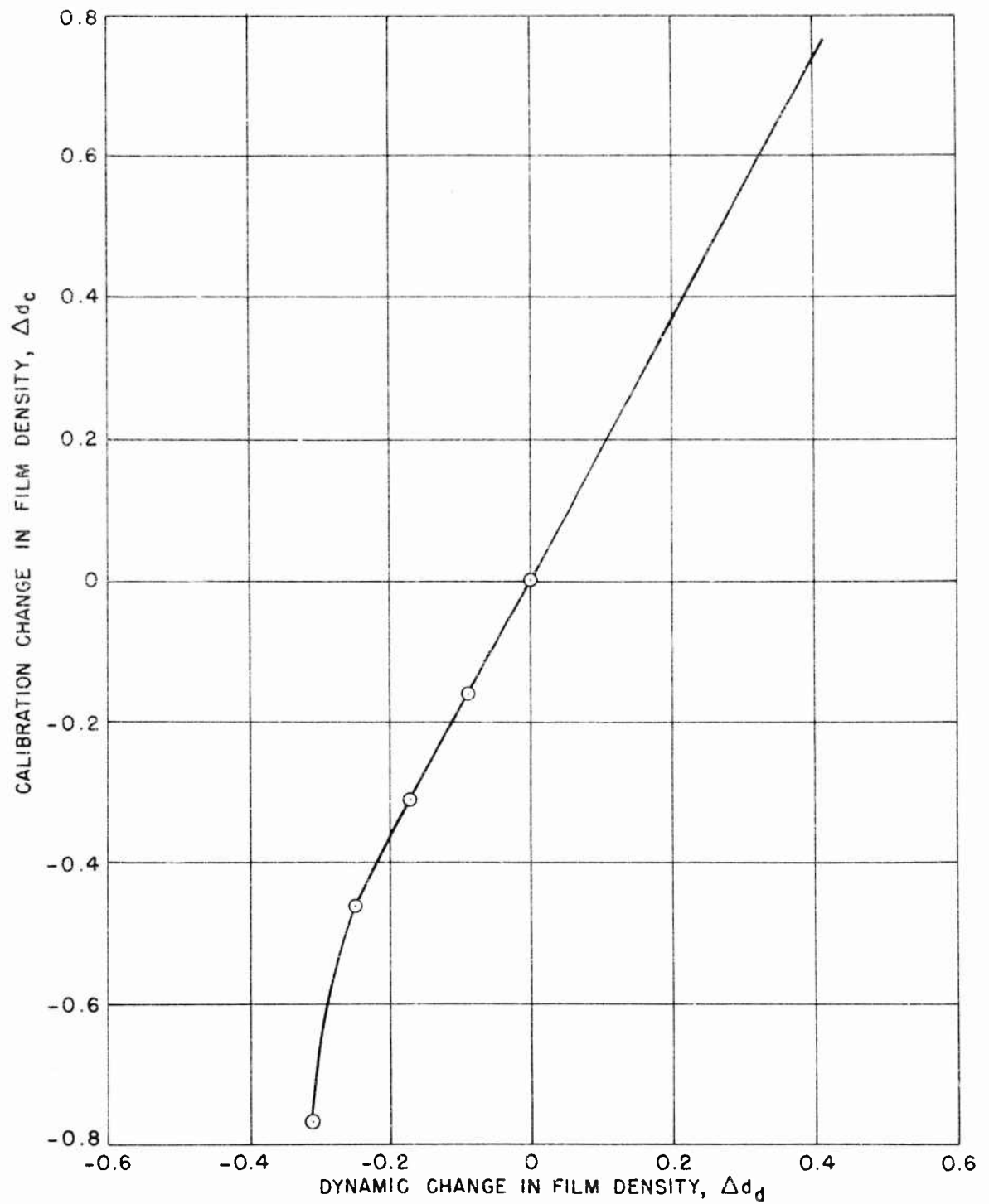


FIG. 4 CALIBRATION CURVE

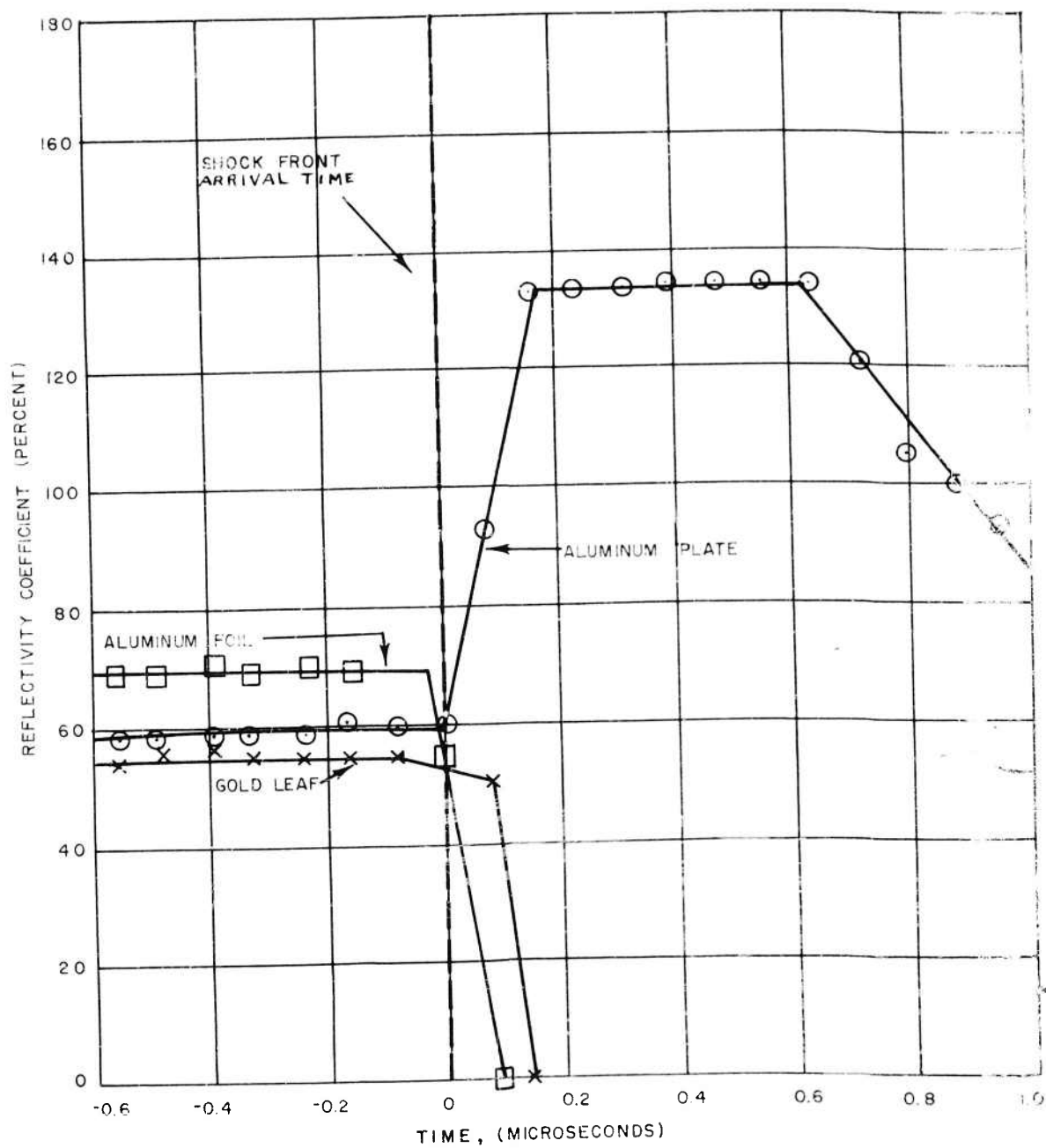


FIG. 5 REFLECTIVITY COEFFICIENT AS A FUNCTION OF SHOCK WAVE ARRIVAL TIME