Investigation of the Photoconductive Effect in Lead Sulfide Films Using Hall and Resistivity Measurements*

JOSEPH F. WOODS
United States Naval Ordnance Laboratory, White Oak, Maryland, and The Catholic University of America, Washington, D. C.
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A long-standing question in the theory of photoconductivity in semiconductor films is whether the change in conductivity can be attributed completely to a change in carrier density, or whether current amplification occurs through modulation of barrier potentials. This question is studied in chemically deposited films of lead sulfide by measuring the fractional change in Hall coefficient, $\Delta R_H/R_H$, and the fractional change in resistivity, $\Delta\rho/\rho$, under illumination. The measurements were made over the temperature range $-41°$C to $+31°$C. It was found that $\Delta R_H/R_H=(\Delta\rho/\rho)(1\pm0.06)$ in this temperature range. Expressed in terms of the Hall mobility, $\mu_H=R_H/\rho$, the result is that $\Delta\mu_H/\mu_H=0\pm0.06\Delta\sigma/\sigma$. A model of photoconductivity is considered, which assumes that the primary photoeffect is a change in the density of majority carriers in the PbS crystallites and that secondary barrier amplification effects can occur by trapping of minority carriers at the intercrystalline barriers. Application of our result, $\Delta\mu_H/\mu_H=0$, to this model leads to the conclusion that photoconductivity in chemically deposited lead sulfide films can be accounted for by changes in majority carrier concentration in the PbS crystallites; no barrier amplification occurs.

I. INTRODUCTION

THE lead sulfide photoconductive film is composed of a system of PbS crystallites, separated by intercrystalline barriers which are probably an oxide of Pb or PbS. The crystallites are about 10^3 to 10^4 angstroms on a side and the width of the intercrystalline barrier is much smaller than this.[1] Space charge regions probably are present at the surfaces of the crystallites, particularly in evaporated films where the oxygen sensitization converts a film from *n*- to *p*-type.[2,3]

In such a complex system it is clear that resistance measurements by themselves cannot be interpreted unambiguously, since resistance is strongly affected by barriers of any kind. The Hall effect, on the other hand, has been shown to be a function of the carrier densities in the crystallites when the intercrystalline barriers are of high resistance compared to the crystallites.[4]

A long-standing question in the theory of photoconductivity[5] in semiconductor films is whether the change in resistance can be attributed entirely to a change in carrier density,[6] or whether current amplification occurs through some kind of barrier modulation.[2,7,8] This problem can be studied by obtaining measurements of the change in resistivity and Hall coefficient (carrier density) under illumination.

This type of measurement requires much greater film stability than do ordinary Hall measurements since one is now interested in small changes. Previous efforts to make such measurements[9] on evaporated films have not led to conclusive results because of instability, noise, and nonuniformity of the films. They have, however, indicated that under strong illumination the fractional change in Hall coefficient is roughly equal to the fractional change in resistivity.

The chemically deposited PbS cells manufactured by Eastman Kodak Company should be more satisfactory than evaporated films for this type of measurement. Lummis and Petritz[10] found them sufficiently free from $1/f$ noise to permit the observation of semiconductor generation-recombination noise. They are quite stable and relatively reproducible in sensitivity, time constant, and responsivity. The photoconductive response is macroscopically uniform over the surface of a film although optical scanning with beams of the order of a few microns in diameter[11] has shown a fine structure.

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[5] For a review of photoconductivity and extensive bibliographies see: T. S. Moss, Proc. Inst. Radio Engrs. **43**, 1869 (1955); R. A. Smith, in *Advances in Physics* (Taylor and Francis, Ltd., London, 1953), Vol. 2, p. 321; and T. S. Moss, *Photoconductivity* (Butterworths Sciencific Publications, London, 1952), p. 37.

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[9] J. W. Lothrup, thesis, Northwestern University, Evanston, 1949 (unpublished); J. L. Levy, Phys. Rev. **92**, 215 (1953).

[10] F. L. Lummis and R. L. Petritz, Phys. Rev. **86**, 660(A) (1952); F. L. Lummis and R. L. Petritz, Phys. Rev. **105**, 502 (1957).

[11] D. Dutton, in *Proceedings of the Conference on Photoconductivity, Atlantic City, 1954* (John Wiley and Sons, Inc., New York, 1956), p. 591.

In this paper we report measurements of the change in the Hall coefficient and resistivity on these films under illumination.[12] The results are used to discuss the relative importance of changes in carrier concentration as compared to barrier modulation in the photoconductive process.

II. APPARATUS AND PROCEDURE

The films were deposited on glass slides with sensitive areas 2.6×1 cm^2. Electrical contacts were evaporated gold. The current leads extended the full width (1 cm) of the film, and the Hall and resistivity electrodes were dots 0.17 cm in diameter. The two Hall electrodes were centered on the long sides of the sensitive area and the four resistivity electrodes were equidistant between the Hall electrodes and the current contacts. The configuration is shown in Fig. 1(a). The films were enclosed in a dry nitrogen atmosphere in order to minimize the effects of ambient changes. The brass container is shown in Fig. 1(b).

In order to verify that the films were uniform, the photoconductive changes in various portions of the film were compared to the total photoconductive change of the film. Under uniform illumination the fractional resistance change of the whole film was measured between the current electrodes and compared with the change measured (1) between the Hall electrodes and (2) between each of the four resistivity electrodes and the adjacent Hall electrode [pairs *AB*, *BC*, *DE*, *EF* in Fig. 1(a)]. All values agreed within 6%. It was concluded that the cells were sufficiently uniform for the purposes of this study.

Hall measurements were made from 31°C to −78°C. Due to noise and thermal drift, however, few measurements were made at temperatures below −40°C. The illumination was varied by controlling the current through a tungsten filament lamp. At room temperature a system of lenses and mirrors furnished a high level of illumination which was quite uniform over the film area. When the cell was mounted in a Dewar flask for cooling, illumination was somewhat nonuniform over the area of the film. The effect of the nonuniformity was determined by observing the difference between room temperature measurements made with the cell mounted for cooling and those made with uniform illumination on the same film.

TABLE I. Typical measurements on a PbS chemical cell in the dark at various temperatures (thickness≅0.5 micron).

	24°C	−39°C	−78°C
Bias voltage (volts)	6	18	45
Bias current, I (microamperes)	3.8	0.47	0.15
Hall voltage, V_H (microvolts)	260	540	640
Hall coefficient, R_H (cm^3/coulomb)	+140	+2340	+8800
Resistivity, ρ (ohm-cm)	29.8	723	5600
Hall mobility, μ_H (cm^2/volt-sec)	4.7	3.24	1.57

The Hall voltage was compared with a calibration voltage applied in series with the Hall electrodes. Figure 1(c) shows the circuit used when the film resistance was expected to exceed 20 megohms during a series of measurements. Both the Hall voltage and the calibration voltage were amplified by an Applied Physics Corporation Model 30 Vibrating Reed Electrometer (*D* in figure) and recorded on a Brown strip-chart 5-millivolt recorder (*E*). This combination had a sensitivity of about 5 microvolts per chart division (1.4 mm) for film resistances up to 10^{11} ohms. The film resistance was measured with a Wheatstone Bridge circuit using a Leeds and Northrup Model 9836 indicating dc amplifier (*B*) as null detector.

When the film resistance was less than 20 megohms the Wheatstone Bridge circuit was not used; the film resistance was computed from the dc current-voltage relation. The Leeds and Northrup amplifier was used as the signal amplifier, with a capacitor and resistor in series with the input to filter out the probe unbalance and slow drift, thus making it unnecessary to make a dc balance. A time constant RC of about 20 seconds was used. This circuit had a higher voltage sensitivity than that of Fig. 1(c) for film resistances less than 7×10^5 ohms and was quicker and simpler to use However, the sensitivity dropped sharply at film resistances above 20 megohms. Measurements made with the two circuits agreed within the accuracy of the experiment.

The magnetic field was uniform within one percent over the area of the film. The magnet current was monitored throughout the measurements to about one percent. The Hall coefficient was constant for a current range of 0.5 to 50 microamperes and for field strengths from 500 to 4500 gauss.

In the presence of noise and drift it was found possible to improve the accuracy of measurement by using a 0.05-cps square wave magnetic field and calibration signal. The magnet used had an inductance of about 0.5 henry and a resistance of about 10 ohms, giving a time constant much shorter than the period of the wave. The response times of the amplifiers and recorder were such that the amplitude of the recorder trace was reduced about five percent by switching. However, the proportionality between Hall voltage and calibration voltage was not affected.

With current flowing in the film a calibration signal was recorded for 3 to 6 cycles. The magnetic field was then applied and the Hall voltage recorded for 6 cycles or more as needed. When noise was appreciable, a greater number of cycles improved the accuracy of the averages. A sample recorder trace of calibration signal and Hall voltage is shown in Fig. 1(d).

The film current or resistance was read before and after the Hall voltage was recorded, and the film and magnet currents were monitored during the recording of the calibration and Hall signals. This procedure

[12] Preliminary reports are: J. F. Woods, in *Proceedings of the Conference on Photoconductivity, Atlantic City, 1954* (John Wiley and Sons, Inc., New York, 1956), p. 636; and Phys. Rev. **99**, 658(A) (1955).

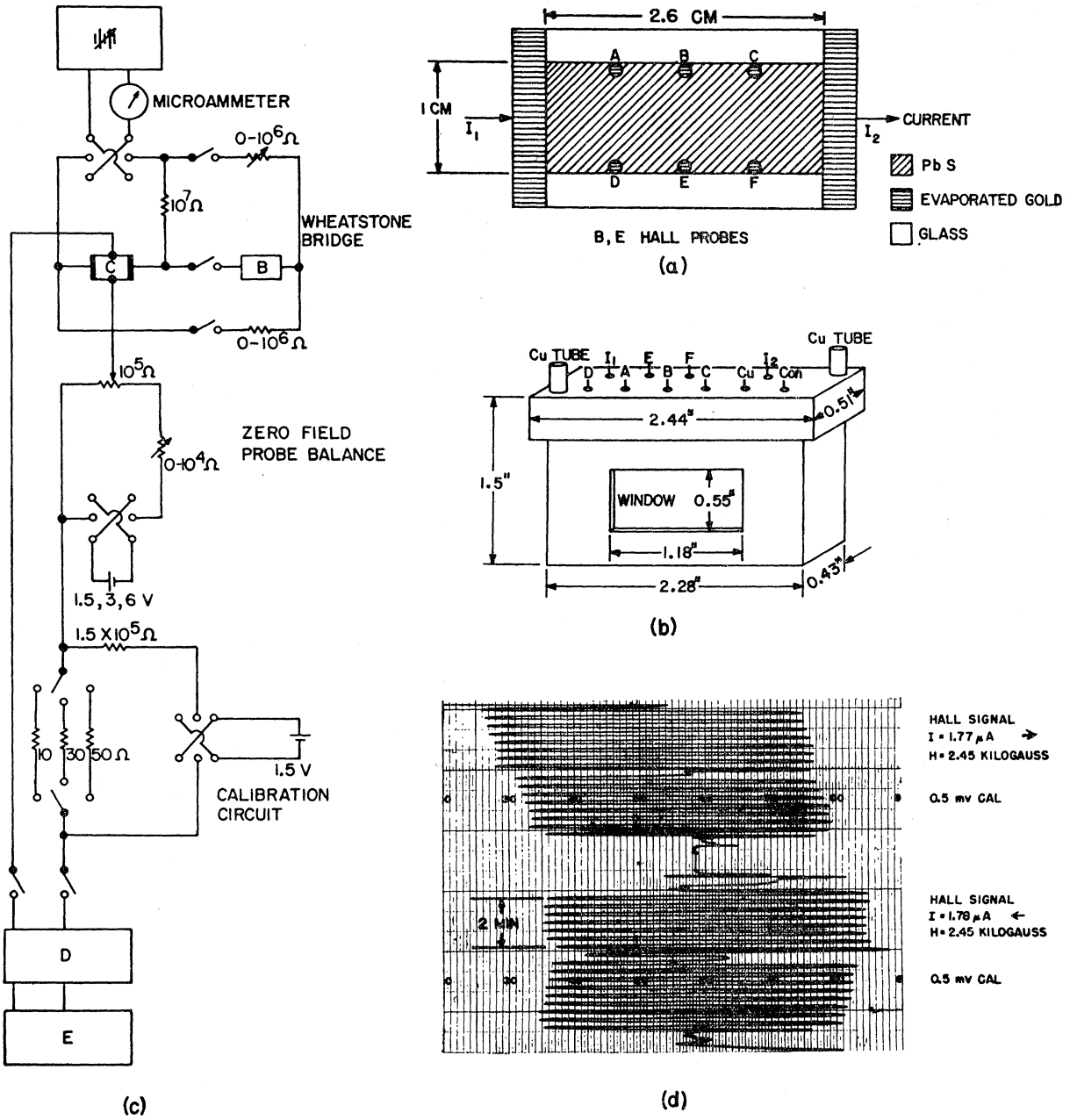

FIG. 1. (a) PbS cell showing electrode geometry. (b) Air-tight cell container with window to admit radiation. (c) Circuit diagram for cell impedances exceeding 20 megohms. Capital letters identify instruments as follows: *B*—Leeds and Northrup indicating dc amplifier as null detector, *C*—PbS cell, *D*—vibrating reed electrometer (10^{12} ohms input resistance) as Hall voltage detector, *E*—self-balancing potentiometer recorder. (d) Sample trace of Hall voltage and calibration signal.

was then repeated with the film current reversed. The total time required was about 10 minutes.

III. RESULTS

A set of typical values observed in these measurements is given in Table I. The values of the Hall coefficient obtained from various films are in substantial agreement, using the value 0.5 micron for the thickness in each case. This value was measured on one film by means of a Michelson interferometer. Since the films are produced by a closely controlled procedure, this thickness was assumed for all the cells. The equilibrium dark values of Hall coefficient (R_H), resistivity (ρ), and Hall mobility[13] ($\mu_H = R_H/\rho$) for 4 cells are listed in Table II.

In the course of continuous measurements using

[13] It should be noted that, since ρ is determined largely by intercrystalline barriers and R_H by the properties of the crystallites, μ_H is not a true mobility. This may be seen from Eqs. (15) and (17). The point is discussed in reference 8.

TABLE II. Properties of PbS chemical cells at room temperature in the dark.

Cell	Ambient	R_H(cm³/coulomb)[a]	ρ(ohm-cm)	$\mu_H = R_H/\rho$ (cm²/volt-sec)
1*A*	air	+121	38.3	3.16
2*A*	air	+148	31.3	4.73
1*B*	air	+146	28.3	5.16
1*B*	N_2[b]	+695	66.5	10.4
2*B*	N_2	+133	25.9	5.13

[a] W. W. Lawrence, formerly of U. S. Naval Ordnance Test Station, China Lake, California, has found dark values of R of +130 and +160 cm³/coulomb on two Eastman films. (Private communication.) His measurements were made on an ac Hall effect system operating at 30 cps.
[b] Cell 1*B* was changed, apparently by a combination of heating and pumping, while being sealed in N_2 atmosphere.

illumination and cooling, the dark values of R_H and ρ changed slowly (sometimes increasing and sometimes decreasing) over the course of days, returning to the values listed in Table II as equilibrium values. The Hall mobility did not change appreciably, nor was the relation between the fractional changes of R and ρ under illumination affected by this slow variation in the dark values.

Figure 2 is a plot of the fractional change of the Hall coefficient, $\Delta R_H/R_H$, *versus* the fractional change in resistivity, $\Delta\rho/\rho$. The data were obtained over a period of nine months on two cells; one at room temperature, the other at temperatures between +31°C and −41°C. The data were obtained with various degrees of uniformity of illumination. This figure illustrates both the trend of data and the deviations caused by lack of uniformity in the illumination.

We want to determine the functional relation between $\Delta R_H/R_H$ and $\Delta\rho/\rho$. From Fig. 2 we see that we can write as a first approximation to this relation,

$$\Delta R_H/R_H = m\Delta\rho/\rho, \quad m \cong 1, \tag{1}$$

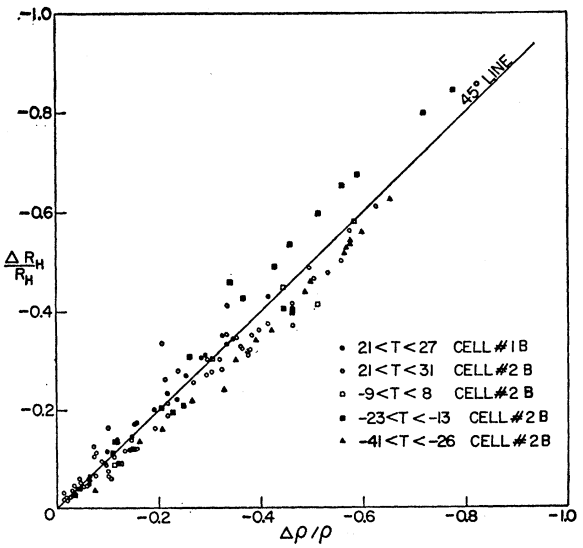

FIG. 2. Compilation of all observations of $\Delta R_H/R_H$ *vs* $\Delta\rho/\rho$ under illumination taken over a nine-month period at various temperatures and with varying degrees of uniformity of illumination. Cell Nos. 1*B* and 2*B*.

where the factor m must be more precisely determined. In order to do this we examine data obtained on a single cell under well-defined conditions of illumination.

Figure 3 is a plot of $m=(\Delta R_H/R_H)/(\Delta\rho/\rho)$ at room temperature, obtained on one cell with low noise and uniform illumination. The position of the light source was not changed during the course of the measurements. For values of $\Delta\sigma/\sigma<1.0$, the error limits indicated are of a statistical origin and will be discussed in the next section. For $\Delta\sigma/\sigma>1$, the indicated limits are due to heating of the film. Observations of R_H and ρ (without illumination) as a function of temperature (Table I and the paper of Petritz *et al.*[14]) showed that the ratio R_H/ρ decreased as T decreased. That is, $\Delta\rho/\rho$ due to heating is greater in absolute value than $\Delta R_H/R_H$ due to heating. Therefore, illumination of sufficient intensity to cause heating will lead to values of $(\Delta R_H/R_H)/(\Delta\rho/\rho)$ less than the value due to the photoconductive process alone. Illumination which increased σ by a factor of 3 at room temperature caused a temperature increase of several degrees in the

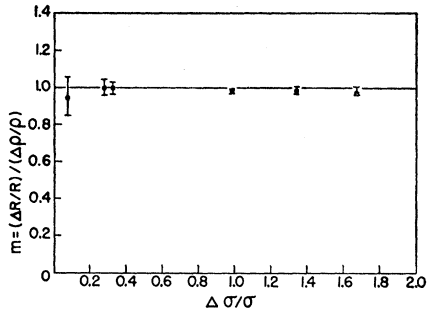

FIG. 3. $(\Delta R_H/R_H)/(\Delta\rho/\rho)$ *vs* $\Delta\sigma/\sigma$ under conditions of uniform illumination and low noise at room temperature. Cell No. 2*B*.

course of a measurement. For this reason, measurements with $\Delta\sigma/\sigma>2$ are not presented.

The data of Fig. 4 were obtained on the same cell as those of Fig. 3 over a range of temperatures. Because of the Dewar flask, the position of the light was necessarily different from that in Fig. 3, and the illumination was nonuniform. This position was roughly the same for all points of Fig. 4. The mean value of $(\Delta R_H/R_H)/(\Delta\rho/\rho)$ is 0.9 at all temperatures. From Fig. 4 we conclude that the relationship which holds between $\Delta R_H/R_H$ and $\Delta\rho/\rho$ at room temperature also holds at low temperatures. Furthermore the only difference between the experimental conditions of the room temperature curves of Fig. 3 and those of Fig. 4 is the uniformity of illumination. We therefore conclude that under illumination, Eq. (1) holds over the range of illumination and temperature studied.

Another way of expressing this result is in terms of the Hall mobility, μ_H, defined as

$$\mu_H = R_H/\rho. \tag{2}$$

[14] Petritz, Lummis, Sorrows, and Woods, in *Semiconductor Surface Physics* (University of Pennsylvania Press, Philadelphia, 1957), p. 229.

From Eq. (2) we find

$$\frac{\Delta\mu_H}{\mu_H}=\frac{(\Delta R_H/R_H)-\Delta\rho/\rho}{1+\Delta\rho/\rho}. \tag{3}$$

Substituting Eq. (1) into Eq. (3), we find

$$\Delta\mu_H/\mu_H\cong 0; \tag{4}$$

i.e., the Hall mobility remains constant under illumination.

IV. ACCURACY AND RELIABILITY OF RESULTS

The large chart of the recorder makes it easy to read V_H to 0.5% since the small divisions are 0.5% of the full scale [see Fig. 1(c)]. This was approximately equal to the over-all resolution of the apparatus. However, the current noise was usually at least 1 to 3% of the Hall signal voltage. Therefore, to obtain the 0.5% accuracy, one would have to average many reversals of magnetic field. For low noise situations it

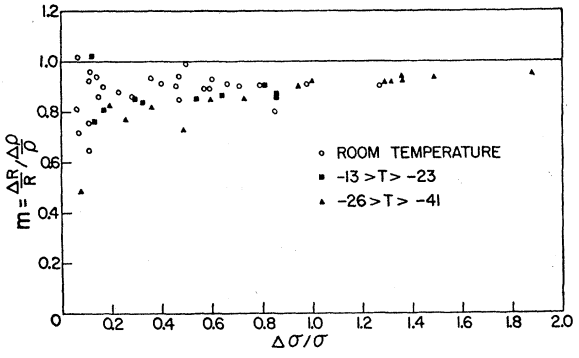

FIG. 4. $(\Delta R_H/R_H)/(\Delta\rho/\rho)$ *vs* $\Delta\sigma/\sigma$ under slightly nonuniform illumination. Cell No. 2*B*. The illumination geometry is the same for all three temperature ranges.

was found that six cycles were sufficient to obtain about 1% accuracy [see Fig. 1(c)]. When the noise was higher a larger number of cycles was necessary. The conductivity and bias current were also measured to better than 1% accuracy.

We can thus evaluate the errors in the parameters of interest, namely dm/m, R_H, ρ, and μ_H. It can be shown that under the conditions of the experiment the measured values of $\Delta R_H/R_H$, $\Delta\rho/\rho$, and $\Delta\mu_H/\mu_H$ are accurate to within ± 0.02, nearly independent of the level of illumination. The error in m, however, is given by

$$dm/m=(2dV_H/V_H)/(\Delta\sigma/\sigma). \tag{5}$$

This relation, together with the voltage accuracy

$$dV_H/V_H=0.01, \tag{6}$$

establishes the lower limit of $\Delta\sigma/\sigma$ which will yield any given degree of accuracy in m. For example, to obtain dm/m accurate to 5% one needs illumination sufficient to yield $\Delta\sigma/\sigma=0.4$. The data of Fig. 4 show

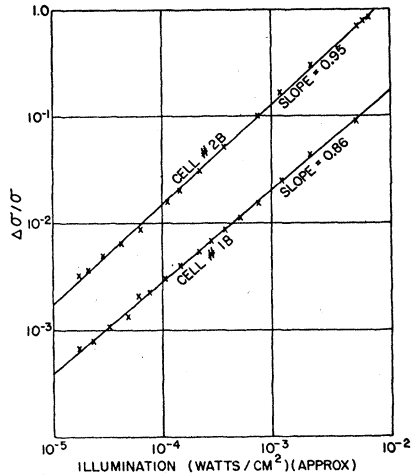

FIG. 5. Cell response $\Delta\sigma/\sigma$ *vs* radiation intensity. Cell Nos. 1*B* and 2*B*.

that dm/m increases with decreasing values of $\Delta\sigma/\sigma$, as predicted by Eq. (5). It would be desirable to make observations at much smaller values of $\Delta\sigma/\sigma$, since PbS photocells are often used with $\Delta\sigma/\sigma$ as small as 10^{-3} to 10^{-5}. However, dm/m would then be 20 to 2000.

In order to determine whether the results can be applied to the region of very small changes, $\Delta\sigma/\sigma \ll 0.2$, where direct measurements of $\Delta R_H/R_H$ are not possible, the dependence of $\Delta\sigma/\sigma$ on illumination intensity was measured over a range which overlapped the region of low-level illumination and the levels used in the Hall effect measurements. A point-source lamp at a variable distance from the cell was used; the intensity was calculated by using the inverse square law. The results are shown in Fig. 5 for two cells. Since there is no deviation from a straight line in either plot over the range of observation, one may conclude that there is no change in the photoconductive mechanism in this range and that the observed relation between $\Delta R_H/R_H$ and $\Delta\rho/\rho$ will hold in the region of small changes, i.e., that m does not change over the region $10^{-5}<\Delta\sigma/\sigma<2$.

Therefore, we can evaluate m at an optimum condition, considering statistical and systematic errors (nonuniform illumination and heating). At $\Delta\sigma/\sigma=0.35$, we find from Fig. 3 that

$$m=1. \tag{7}$$

Equation (5) indicates that at this point

$$dm/m\cong 0.06. \tag{8}$$

Higher values of $\Delta\sigma/\sigma$ could be used to reduce the statistical error in m, but would begin to introduce a systematic heating error.

Substituting Eqs. (7), (8), and (3) into Eq. (1), we find

$$\Delta R_H/R_H=(\Delta\rho/\rho)(1\pm 0.06), \tag{9}$$

$$\Delta\mu_H/\mu_H=0\pm 0.06\Delta\sigma/\sigma. \tag{10}$$

V. APPLICATION OF RESULTS TO THEORETICAL MODELS OF PHOTOCONDUCTIVITY

To discuss the above results [Eqs. (9) and (10)] in terms of a given model, one must derive expressions for R_H, ρ, and μ_H, and for changes in these quantities under illumination. We shall not attempt to do this systematically for all models in the literature, but shall discuss only a single model[8] which has been successful in correlating experimental measurements of noise,[10] responsivity, and sensitivity[15] with theoretical values. We shall require the following properties of the model: (1) the crystallites are assumed to be sufficiently homogeneous and similar that an average carrier density, p, is representative of the whole film; (2) the intercrystalline barrier regions are thin and of high resistivity compared to the crystallites; (3) the primary photoeffect, a change in the majority carrier density, results from absorption of light in the PbS crystallites, creating hole-electron pairs; (4) secondary amplification effects can result from lowering intercrystalline barriers by trapping minority carriers; and (5) space charge effects within the crystallites are neglected.

The current density across a single barrier is given by[16]:

$$j=pMe^{-q\phi/kT}(e^{q\Delta V_b/kT}-1), \tag{11}$$

where p is the density of holes in the crystallites adjacent to the barrier, q is the electronic charge, ϕ is the potential (height) of the barrier, ΔV_b is the voltage across the barrier, and M depends on the particular barrier theory used and is independent of p and ϕ. Because of the many barriers in the film, $q\Delta V_b/kT\ll 1$ for normally applied voltages and the barrier is ohmic; thus

$$j=pMe^{-q\phi/kT}q\Delta V_b/kT. \tag{12}$$

From Eq. (12) and properties 1, 2, and 5 one can show that the Hall coefficient,[4] macroscopic resistivity,[8] effective mobility,[8] and barrier modulation factor[8] B of the polycrystalline film are, respectively,

$$R_H=3\pi/8qp, \tag{13}$$

$$\rho=1/q\mu^*p, \tag{14}$$

$$\mu^*=(M/nkT)e^{-q\phi/kT}, \tag{15}$$

$$B=(\Delta\mu^*/\mu^*)/(\Delta p/p), \tag{16}$$

where n is the average number of crystallites per centimeter of length of the film. Therefore, from Eqs. (13) and (14) the Hall mobility is directly related to μ^* by

$$\mu_H=R_H/\rho=\tfrac{3}{8}\pi\mu^*, \tag{17}$$

and includes the effect of the barrier potential [Eq. (15)].

When light is absorbed, we have from Eqs. (13)–(17):

$$\frac{\Delta\rho}{\rho}=\frac{(-\Delta p/p)-(\Delta\mu^*/\mu^*)(1+\Delta p/p)}{(1+\Delta\mu^*/\mu^*)(1+\Delta p/p)}, \tag{18}$$

$$\Delta R_H/R_H=-\Delta p/[p(1+\Delta p/p)], \tag{19}$$

$$\Delta\mu^*/\mu^*=e^{-q\Delta\phi/kT}-1=\Delta\mu_H/\mu_H. \tag{20}$$

Our experimental result, Eq. (9), requires the right-hand side of Eq. (18) to equal that of Eq. (19) for arbitrary levels of illumination. For this to be the case, $\Delta\mu^*/\mu^*$ must be zero within experimental error. This result can also be derived by substituting the experimental result of Eq. (10) into Eq. (20).

We therefore conclude that in this model there can be no large change in barrier potential under illumination, that the barrier modulation factor, B, is zero, and that the photoconductive effect must be due entirely to an increase in the density of majority carriers in the PbS crystallites.

VI. SUMMARY

From measurements of the change in resistivity and Hall coefficient under illumination it is found that in chemically deposited lead sulfide films,

$$\Delta R_H/R_H=(\Delta\rho/\rho)(1\pm 0.06), \tag{21}$$

$$\Delta\mu_H/\mu_H=0\pm 0.06\Delta\sigma/\sigma, \tag{22}$$

over the temperature range $+31°$ to $-41°$C, and for illumination levels such that $10^{-5}<\Delta\sigma/\sigma<2$. The establishment of these relations provides an experimental criterion for the selection of an adequate theory of photoconductivity in lead salt semiconducting films.

The relations were used to evaluate the relative amount of barrier amplification occurring in PbS films on the basis of a recently published theory of photoconductivity[8]; it was concluded that no significant barrier amplification can occur. Therefore, the photoconductive effect is entirely due to an increase in the density of majority carriers in the lead sulfide crystallites.

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[16] H. C. Torrey and C. A. Whitmer, *Crystal Rectifiers* (McGraw-Hill Book Company, Inc., New York, 1948), pp. 77–82.