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Examination of Colored Alkali Halides for Photoelectric Hall Effect

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Five additively colored alkali halides have been examined for photoelectric Hall effect by means of two different arrangements, a single cross-electrode set-up and Tartakowsky's divided electrode method. The consistently null results obtained were checked by using zincblende, a substance of known photoelectric galvanomagnetic properties, in both arrangements. From the upper limit assigned to the Hall effect, it was computed that the mean free path of photoelectrons in rocksalt is not much greater than the dimensions of a lattice cell—in agreement with Von Hippel's result for the electronic mean free path on the basis of electrical breakdown experiments. This interpretation of the null result was supported by the fact that the large initial surge of electron current in KCl and KBr at the outset of illumination also showed no Hall effect. The magnetic deflection of photoelectric current reported in rocksalt by Tartakowsky was not found, either for additively or photochemically colored specimens.

POHL[1] and his collaborators have made exhaustive studies upon the photoelectric properties of additively colored alkali halide crystals, but so far have not reached a definite decision as to the nature of the color centers or of the complete conduction mechanism of the photoelectric current. Lukirsky[2] has reported a Hall effect in photochemically colored rocksalt such that the mean free path of the photoelectrons was calculated (according to Joffé[3]) to be "of the order of 10^{-5} or 10^{-6} cm." Tartakowsky[4] has described an apparatus by means of which he demonstrated an "electron polarization" set up in photochemically colored rocksalt by the magnetic deflection of the lines of current flow to a divided collecting electrode. Von Hippel,[5] however, on the basis of experiments upon electrical breakdown in rocksalt, has concluded that the mean free path of electrons therein is of the order of the dimensions of one lattice cell. His conclusion was supported by a theoretical calculation of the "relaxation time" of electrons in rocksalt made by Fröhlich.[6] In view of the discrepancy between Lukirsky's and Von Hippel's results when applied to the determination of electron mean free paths in rocksalt, and also because of the sparseness of available data on photoelectric Hall effects in general, it was considered desirable to examine several colored alkali halides in detail for the photoelectric Hall effect.

APPARATUS

First measurements on NaCl, KCl and KBr were by means of a single cross-electrode arrangement, shown schematically in Fig. 1. The cross electrode was connected directly to the grid *A* of a sensitive direct-current amplifier, with circuit as given by Harnwell and Van Voorhis.[7] A current sensitivity of 4×10^{-15} amp. = 1 scale division was used. B-batteries connected in series to the amount of about 500 volts were applied to the ends of the crystals. These batteries were

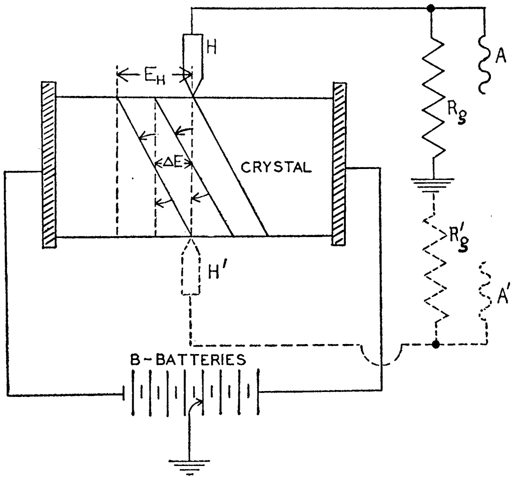

FIG. 1. Schematic showing single cross-electrode method for testing Hall effect in crystal. The rotation of equipotential lines by the Hall field is also shown.

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[1] R. W. Pohl, Proc. Phys. Soc. **49**, supplement, 3 (1937).
[2] P. Lukirsky, J. Russ. Phys. Chem. Soc. 1916 (in Russian).
[3] A. F. Joffé, *The Physics of Crystals* (McGraw-Hill), p. 129.
[4] P. Tartakowsky, Zeits. f. Physik **66**, 830 (1930).
[5] A. von Hippel, J. App. Phys. **8**, 815 (1937).
[6] H. Fröhlich, Proc. Roy. Soc. **A160**, 230 (1937).
[7] G. P. Harnwell and S. N. Van Voorhis, Rev. Sci. Inst. **5**, 244 (1934).

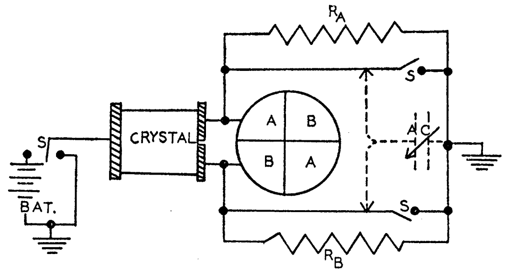

FIG. 2. Differential electrometer method of Tartakowsky. The high resistors R_A and R_B were not part of Tartakowsky's circuit, which included instead the equalizing variable air condenser AC (shown in dashed lines).

grounded near their center, the grounded point G being varied by steps of $1\frac{1}{2}$ volts until no current flowed into H when the crystal was steadily illuminated. Batteries and leads were electrostatically shielded.

Measurements on NaCl, KCl and KBr were repeated by means of Tartakowsky's divided electrode arrangement (Fig. 2). KI and NaBr were also measured with this latter apparatus.

Single crystals of the substances studied were manufactured from chemically-pure, fused salts with an apparatus similar to that described by Walther.[8] Photoelectric contacts with the crystals were made by sputtering mirror-like, platinum coats on to freshly cleaved faces. The small side electrodes were made by scraping away all of the sputtered coat on one side of a specimen except that covered by a clamped safety razor blade (thickness 0.24 mm). Crystals were additively colored by electron impregnation at high temperature (500 to 700°C) and then "quenched" in an air stream to secure atomic dispersion of color centers.

The crystals were gripped between brass electrodes by the steady pressure of spiral brass springs, the electrodes being mounted in a hard rubber block frame. The frame, with glass plates, also served as a low vacuum chamber. The Hall electrode was protected fron leakage currents by a guard ring. Crystals were illuminated over their complete length by the full light from the tungsten filament of a 6-8-volt, 50-cp automobile headlight bulb, the focused light beam being sent through a long hole in one pole piece of the Weiss electromagnet which produced the magnetic field.

PROCEDURE AND RESULTS

The procedure was to adjust the position of the grounded point of the applied batteries until with the light shining on the specimen, no photoelectric current flowed into the cross-electrode. Then a magnetic field was applied normal to the principal plane of the specimen, and any resulting effect upon the current into the Hall electrode observed.

The arrangement was empirically tested for its sensitivity to small e.m.f.'s by shifting the grounded point of the batteries by a small amount and noting the corresponding effect on the *steady* deflection of the indicating instrument. Such a shift had the effect of increasing the potential difference between the Hall electrode and the respective ends of the crystal by an amount $\pm\Delta E$.

It was assumed, from considerations of symmetry, that a transverse Hall e.m.f. E_H across the crystal would rotate the equipotential lines of the electric field about their points of intersection with the longitudinal center of the crystal, i.e. (Fig. 1), that the potential difference between the Hall electrode and the ends of the crystal would be changed by $\pm\frac{1}{2}E_H$, respectively. This assumption was made plausible by an experiment upon zincblende, which from Lenz'[9] work is known to have an easily detectable photoelectric Hall effect. With zincblende in the apparatus it was found that the direction of the Hall current into the single cross-electrode could be reversed by reversing the direction of the applied electric field, indicating that *if* a second cross-electrode H' (Fig. 1) had been connected to a second amplifier A', the latter, during application of a magnetic field, would have recorded a Hall current equal and opposite to that recorded by A. Hence it was assumed that the Hall e.m.f. E_H across the width of a given alkali halide crystal could not be greater than the smallest voltage shift ΔE of the grounded point which would produce a positive effect.

No positive indication of a Hall e.m.f. or associated Hall current was found for the steady photoelectric current in additively colored NaCl, KBr or KCl. In one set of readings upon rocksalt it was found that the *steady* change in the

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[8] H. Walther, Rev. Sci. Inst. **8**, 406 (1937).

[9] H. Lenz, Ann. d. Physik **77**, 449 (1925); **82**, 775 (1927).

reading of the indicating instrument for small shifts of the grounded point was 7.5 cm/volt, whereas turning on the magnetic field (of 12,200 gauss) produced random shifts of about 1.5 cm. The variations apparently caused by application of the magnetic field were found to be present with or without the light shining on the crystal. The average of 8 readings taken with the magnetic field in one direction differed by less than 0.3 cm from the average of 8 readings with the magnetic field reversed. Therefore, assuming that for the very small currents involved any Hall e.m.f. present would be proportional to the transverse photoelectric current produced by it, it was concluded that the effect of *reversing* the magnetic field was less than that produced by shifting the grounded point of the batteries by 0.1 volt. With the calibrating factor of $\frac{1}{2}$, the Hall e.m.f. was less than 0.2 volt across the crystal (dimensions $7.5\times 4.0\times 1.2$ mm), or, the Hall field Y was less than 0.5 volt/cm.

The steady photoelectric current under the applied voltage, 520, was found to build up a photoelectric "back e.m.f." of 90 volts, as determined by the applied potential which would just prevent the built-up back photoelectric e.m.f. from sending a current in the opposite direction. Then the total "effective" potential difference over the length of the crystal was 430 volts, and the average resultant field 570 volts/cm. Therefore the ratio Y/X of the Hall field to the electric field near the center of the crystal was less than 1/1150.

The measurements just described were taken with a very lightly colored specimen, color density estimated at about 5×10^{16} centers/cm^3. According to Glaser and Lehfeldt[10] the so-called "thrust paths" of the photoelectrons in rocksalt are inversely proportional to the color center density. A thrust-path, however, presumably consists of many free paths.

The measurements were checked and confirmed by using the amplifier in a slightly different manner, namely, by allowing the grid to "float" (R_g = insulation leak resistance = about 10^{13} ohms) and noting its rate of increase of potential, if any, caused by application of a magnetic field to the illuminated crystal.

Lenz[9] found the photoelectric Hall effect in zincblende to depend in sign and magnitude upon the orientation of the specimen, while Von Hippel[5] found preferred directions of electron movement in rocksalt. It therefore appeared likely that the Hall effect in rocksalt might be a function of the orientation of the specimen. A plate was cut and sputtered so that the electric field could be applied in a 110-direction, and the possible Hall field measured in another 110-direction. No effect was found, the upper limit to the ratio Y/X for a magnetic field of 12,200 gauss being set at 1/200.

It seemed possible that the small value of the Hall effect indicated for the *steady* photoelectric current was due to opposite and canceling contributions by the equal components of electron and "hole conduction" current, since, according to an application of quantum mechanics by Fowler,[11] the Hall coefficient for an electronic semi-conductor is

$$R=\frac{kY}{HX}=\frac{3}{8}\frac{\sigma_1-\sigma_2}{e(n_1+n_2)},$$

where k = constant, H = magnetic field, e = charge on a carrier, n_1 and n_2 are the numerical concentrations of the negative and positive carriers, respectively, and σ_1 and σ_2 are the corresponding conductivities. Now the colored potassium salts are characterized by a comparatively large initial surge of photoelectric current[1, 10] at the outset of illumination, followed by a rapid decrease to a final "steady" (but still decreasing) value about 100 times smaller than the initial current. The rapid decrease of current is attributed to the formation of space charge, resulting from the failure of the positive component of photoelectric current to remove positive charge as rapidly as the negative charge is drawn out. The initial photoelectric surge therefore consists mostly of electrons, and consequently in it the canceling contribution of the positive carriers to the Hall effect should be small. Therefore measurements were taken of the effect of an applied magnetic field upon the photoelectric current flowing during a 1/10 second flash of light in KCl and KBr, with the amplifier, still connected to the cross-electrode, as a ballistic instrument.

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[10] G. Glaser and W. Lehfeldt, Gott. Nachr. **2**, 7 (1936).

[11] R. H. Fowler, Proc. Roy. Soc. **A140**, 504 (1933).

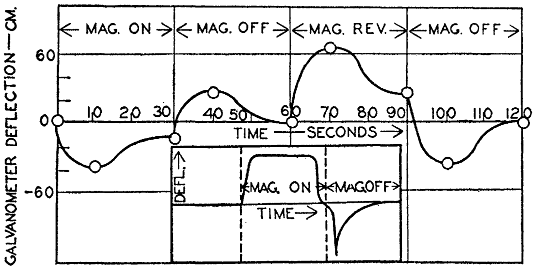

FIG. 3. Upper curve, "photoelectric Hall back e.m.f." effect in ZnS by single cross-electrode set-up. The initial large throw of the indicating instrument upon application of a magnetic field was ascribed to a lack of photoelectric back e.m.f. initially opposing the Hall current; the decrease to a nearly steady value within 30 seconds was ascribed to the formation of a photoelectric back "Hall" e.m.f., whose presence then caused the reverse throw upon release of the magnetic field. Inset lower curve, Tartakowsky's curve for rocksalt, showing a similar "polarization" by means of the divided electrode arrangement.

With the KCl specimen ($7.0\times 3.5\times 1.0$ mm) and intense illumination, the smallest obtainable unidirectional throw of the indicating galvanometer (for 0.1-second flashes) was about 25 cm. (Space charge built up rapidly during the flash, changing the potential distribution along the length of the crystal, so that for some positions of the grounded battery point the direction of the photoelectric current into the Hall electrode reversed during the flash.) Changing the grounded point by 3 volts changed the ballistic throw, on the average, 6 cm, or 2 cm/volt, whereas, for 12 pairs of readings, the average for the magnetic field in one direction did not differ by more than 0.1 cm from the average with the magnetic field reversed. Random fluctuations between consecutive readings were of the order of 1 cm but a persistent effect of $\frac{1}{2}$ cm would have been easily detected. The assumed maximum unobserved effect of $\frac{1}{2}$ cm was due to reversing the magnetic field, so that the effect of applying the magnetic field in one direction must have been less than $\frac{1}{4}$ cm, the effect which according to the calibration would have been produced by a Hall e.m.f. of $\frac{1}{4}$ volt, or a Hall field of 0.7 volt/cm. The upper limit to the ratio Y/X then came out to be 1/850 for $H=12{,}200$ gauss.

However, the photoelectric back e.m.f. developed during the flash was about one-half of the total applied e.m.f., or the resultant time average of the longitudinal electric field near the Hall electrode during the flash was about three-fourths of the initial applied field; likewise with the time average of a possible accompanying Hall field. Making this correction, the limit for ratio Y/X for the initial electron surge of current was taken as $4/3\times 1/850=1/640$.

The crystal was grounded and illuminated for $3\frac{1}{2}$ minutes between consecutive flashes, or long enough to remove at least 99 percent of the developed photoelectric back e.m.f. and to restore the crystal to approximately the same condition for each reading.

In similar measurements upon KBr, in which the photoelectric polarization built up by successive flashes was not completely discharged between consecutive trials, the upper limit to Y/X was 1/225.

To test whether the experimental arrangement was capable of detecting the presence of a small Hall current or e.m.f., a specimen of clear yellow zincblende (sphalerite), $7.27\times 4.8\times 2.0$ mm, cut with its length parallel to a direction symmetrical with respect to the crystal axes (following a suggestion by Lenz[9]), was substituted in the apparatus. A Hall current, whose direction reversed both with reversal of the electric and the magnetic fields, was found to flow upon application of the magnetic field. Data (averages of 20 trials for both directions of the magnetic field) are shown in Fig. 3 for one direction of the electric field.

The change in steady deflection per unit shift of grounded point was 5.7 cm/volt, while the change produced by reversing the magnetic field was 40 cm. From these data, and using the calibration factor of 2, it was found that the Hall field was 14.5 volts/cm, the *average* applied electric field being 890 volts/cm. According to Lenz the electric field near the middle of a photoelectrically polarized zincblende plate is equal, approximately, to the average applied field. Therefore the value, 1/60, found for the ratio Y/X in zincblende subjected to 12,200 gauss, agreed well, at least in order of magnitude, with Lenz' value for the same ratio, 1/44, extrapolated to the same value of magnetic field. (Lenz showed that the Hall field in zincblende is linearly proportional to the strength of the magnetic field.) The difference could be due to structural differences between the specimens of the zincblende, to the assumption made here that the Hall e.m.f. was proportional to the Hall current, or to the fact that Lenz' current densities were about 40 times greater than those used here. In any event it appeared that the single-electrode method was capable of detecting a photoelectric Hall effect.

An effect ascribed to "photoelectric Hall back e.m.f." is shown in the graphical display of the data on zincblende. Upon turning on the magnetic field the galvanometer deflection was about $2\frac{1}{2}$ times greater than its steady deflection. If the magnetic field was then removed, an opposite initial deflection occurred, of the same order of magnitude as the original initial deflection. The original large initial deflection was ascribed to a lack of photoelectric back e.m.f. at the Hall electrode initially opposing the Hall current. The decrease of Hall current was ascribed to the building up of a photoelectric back "Hall" e.m.f., and the reverse current to the action of this built-up e.m.f. after removal of the original Hall field.

A similar effect, obtained by using a divided electrode and a differential electrometer, has been described for x-rayed rocksalt by Tartakowsky.[4] His curve is reproduced as an insert in Fig. 3 for the purpose of comparison. An attempt to duplicate Tartakowsky's curve for rocksalt by means of his set-up was unsuccessful.

The photoelectric back e.m.f. developed by a steady current in KCl was found to be about 90 percent of the total applied e.m.f., and about 99 percent in KBr. Potential measurements made with a movable side electrode showed that the developed back e.m.f. was mostly due to charges collected near the ends of a crystal. The potential distribution along a photoelectrically "excited" KBr crystal is shown in Fig. 4. The irregularity of individual points is a result of the procedure of taking the crystal-holder out of the apparatus between readings in order to move the probe.

The potential distribution resembles closely that found in a gaseous discharge tube. The electric field intensity is greatest close to the electrodes and comparatively small near the middle of the specimen. It might therefore be expected that the Hall field generated by a magnetic field would also be greater near the ends of such a crystal, so that a method for detecting such a possible concentration of Hall field near the electrodes was needed.

According to Lenz the potential distribution along a photoelectrically polarized zincblende crystal is such as to indicate a resultant positive space charge throughout. The electric field intensity, in accordance with Poisson's equation, is increased near the cathode and diminished near the anode. In the present investigation, zincblende was placed in the divided electrode arrangement of Tartakowsky. It was then found that the magnetic deflection of current from one part of the divided electrode to the other was approximately 12 percent greater when the divided electrode was used as cathode than when used as anode. The difference was about ten times greater than the average random fluctuation between consecutive readings. It therefore appeared that the Tartakowsky arrangement (first set up for the purpose of checking Tartakowsky's experiment with colored rocksalt) should be suitable for the detection of small Hall fields concentrated near the ends of a specimen. (It was ascertained that the zincblende specimen as a whole had no rectifying effect upon the photoelectric current.)

Check measurements upon NaCl, KCl and KBr were made with the divided electrode arrangement, as well as measurements upon NaBr and KI. No *conclusive* evidence of a Hall deflection of current was found in any of these substances. Upper limits to the possible Hall fields in the various substances were assigned by direct comparison with the behavior

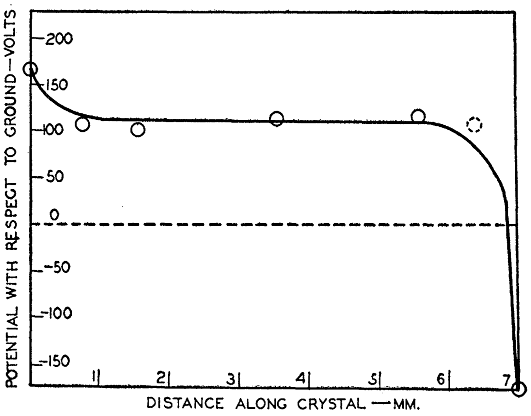

FIG. 4. Potential distribution along photoelectrically polarized KBr crystals, as determined by a movable probe.

of zincblende in the same apparatus. In Table I, i is the total (average) current to one segment of the divided electrode, $\Delta_h i$ is the change of current to that segment (average of 10 readings) caused by the magnetic field, and $\Delta_f i$ is the average fluctuation in the current for a single observation from its average value i. In the last column, R is the effect of the magnetic field upon the current flow in a given alkali halide as compared to its effect upon zincblende. This ratio has been assigned the positive sign if the apparent (very small) current deflection indicated by the average of 10 readings was in the same direction as for zincblende.

There is probably no significance to the fact that the very small magnetic deflection (if any) in KCl, KBr, and NaBr had the correct sign for a normal Hall deflection of current, because for these the average fractional change of current to one part of the divided electrode after application of the magnetic field was several times smaller than the average fractional fluctuation of current between consecutive readings. In rocksalt, however, a small "anomaly," slightly larger than the average fluctuations, and of the correct sign for a normal Hall current, was found. The anomaly was not recognized as an indication of a bona fide Hall effect for the following reasons:

1. In 6 specimens tested, the anomaly had the "wrong" sign in one, and did not exist in another (which happened to be a plate with non-uniform coloring).
2. Most of the effect was found to be produced by application of the magnetic field in *one* direction; the other direction of field had no effect, or even a small effect in the "wrong" direction.
3. Similar effects, but several times smaller, were sometimes observed with the crystal unilluminated.

A possibility remains, however, that this anomaly was an indication of a small Hall effect on the borderline of observational limit, superimposed upon a small, spurious, unidirectional effect of the magnet or magnetizing circuit. The effect was too small to be observed with the original "balanced-rate-of-charge" method of Tartakowsky; it was only observed with the steady deflection method, using high resistors to impress potential differences upon the differential electrometer (Fig. 2).

TABLE I.

Substance	Specimen Size (mm)	$\Delta_h{}^{i/i}$	$\Delta_f{}^{i/i}$	R
NaCl	6.7×4.6	1/880	±1/1000	1/22
NaBr	4.4×3.6	1/4000	±1/1500	1/20
KCl	5.8×4.5	1/1700	±1/600	1/50
KBr	7.5×4.5	1/2300	±1/1000	1/50
KI	3.7×3.5	±1/5000	±1/2000	±1/40

The differential electrometer could also be used to measure the difference in charge collected by the two parts of the divided electrode as a function of the applied magnetic field during a short flash of light. It was found that the charge collected by one electrode-part was changed by less than 1 part in 800 for KCl, 1 in 225 for KBr, and 1 in 600 for KI, by application of a magnetic field of 12,200 gauss, lending further confirmation to the previous null results.

Null results thus far described were obtained with *additively* colored specimens, having a stoichiometric excess of alkali atoms. Lukirsky's and Tartakowsky's reported positive results had been with *photochemically* colored rocksalt. It seemed possible that the discrepancy was due to the difference in the method of preparation of the specimens. Rocksalt colored by exposure to 50-kv tungsten x-rays was therefore tested in the Tartakowsky set up, using the balanced rate-of-change method.

An unsuspected difficulty was immediately encountered—namely, that the x-rayed crystals faded so fast under the fairly bright illumination needed to give "steady" currents of sufficient magnitude that it was hard to make the adjustment of the equalizing condenser before all the photo-sensitivity had disappeared; also it was impossible to obtain a steady state of photoelectric polarization. A fairly dense coloring would last only about five minutes. However by resensitizing the crystal several times, and by making small adjustments between consecutive pairs of readings (one reading for each direction of the magnetic field) so that little photoelectric sensitivity of a specimen was "wasted" and the adjustment constantly improved, sufficient data was finally accumulated. The conclusion was that no persistent effect greater than 1/10 of the corresponding effect observed in zincblende under the same conditions existed in rocksalt colored by x-rays.

The electrometer used had almost exactly the same sensitivity as that of Tartakowsky's instrument. The working of the arrangement was tested with zincblende. The discrepancy is hard to explain. Tartakowsky, however, used a magnetic field of 30,000 gauss, compared to 12,200 gauss used here.

Besides being tested for possible photoelectric Hall effect, NaCl, KBr, and KCl, and also zincblende were examined for possible changes in photoelectric resistance caused by a magnetic field. Fluctuations in the longitudinal photoelectric current itself were found to limit the sensitivity to the extent that the null result uniformly found indicated only that large magneto-resistance anomalies do not exist in the photo-conductors studied. Upper limits to the magneto-resistance change caused by a field of 12,200 gauss were set as follows: NaCl, 0.15 percent; KCl, 0.3 percent; KBr, 0.25 percent; ZnS, 0.14 percent. The maximum expected effect in ZnS, according to calculations based upon the photoelectron mean free path as deduced from Lenz' Hall effect measurements, was only 0.05 percent.

The fluctuations of photoelectric current were five to ten times greater than those which would be expected from shot and Johnson effects[12, 13]. The fluctuations apparently were due to some phenomenon in the crystals. They were observed, not only with the direct-current amplifier, but also with the differential electrometer, which, when electrically shielded, is less sensitive to random ionization of the air about it than the amplifier. The ratio of the fluctuations in current to the entire current increased with increasing applied voltage. In KBr, for example, the fluctuations were about 50 percent of the total current when 600 volts were applied to a crystal 7.2 mm long.

Conclusion and Discussion

It has been established, by means of two different experimental arrangements, both checked with a substance of known properties, that the photoelectric Hall field in additively colored rocksalt, if it exists at all, is less than 1/1000 of the applied electric field (for $H=12{,}200$). Similar upper limits have been obtained for four other colored alkali halides. If, to take the particularly important case of rocksalt, the null result is interpreted as meaning that the free paths of the photoelectrons in the crystal lattice are comparatively short, the equation of Gans,[14]

$$\frac{Y}{X}=\frac{1}{4}\left(\frac{3\pi m}{kT}\right)^{\frac{1}{2}}\frac{e}{m}\lambda H,$$

for the *isothermal* Hall effect in an electronic conductor could be used to determine the order of magnitude of the maximum mean free path (λ) consistent with the experimentally determined upper limit of the ratio Y/X. Putting $Y/X=1/1000$ and $H=12{,}000$ gauss in the foregoing equation, it is found that

$$\lambda=4.5\times10^{-8}\text{cm}.$$

Such a length is not much greater than the dimensions of a lattice cell. Von Hippel, from his experiments on electrical breakdown in rocksalt, concluded that the electronic mean free path was of the order 2×10^{-8} cm. Such a mean free path, by Gans' formula, would give a Hall field, for 12,000 gauss, of

$$Y/X=1/2250.$$

Detection of a Hall effect of such a magnitude was slightly beyond the sensitivity of the measurements which have been described.

An alternative interpretation, that the contribution of the photoelectrons to the Hall field in colored alkali halides is nearly or entirely canceled by the contribution of the positively-charged carriers, might be made, but appears unnecessary in the view of the fact that no Hall effect was found even in the comparatively large initial surge of photoelectric current in KCl, KBr, and KI, this initial surge being, in all probability, almost entirely composed of electrons.

The writer wishes to acknowledge his indebtedness to Professor C. W. Heaps for suggestion of the problem, as well as for many helpful discussions concerning it; and to Professor H. A. Wilson for his continued interest in the investigation and valuable advice in the design of apparatus.

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[14] R. Gans, Ann. d. Physik **20**, 293 (1906).