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Electronic Hall Effect in Diamond*

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A method for measuring the Hall mobility in insulating photoconductors is described which avoids the space-charge difficulties present in previous measurements. Unconventional electrodes are used, and the method differs from the conventional one in several other ways. It is a transient method, and the electric field in the sample is rotated by the external electrodes, rather than by the electrons under study. The direction of charge flow is deduced from a current measurement, rather than by confining the current to a thin sample whose orientation gives the direction of charge flow. No attempt is made to inject or eject electrons through the surface of the sample.

Measurements using this technique have been made on a number of diamonds. Both electrons and holes are found to contribute to the photoconductivity, making interpretation of the data difficult. It is tentatively concluded that the electronic mobility varies as $T^{-\frac{3}{2}}$ and is about 1800 cm^2/volt-sec at 300°K. For holes it is concluded (with far less certainty) that the mobility also varies as $T^{-\frac{3}{2}}$ and is somewhat greater than 1200 cm^2/volt-sec at 300°K. These observations are consistent with the deformation potential theory of electronic mobility in nonpolar crystals.

A. INTRODUCTION

PREVIOUS attempts[1-3] to measure the Hall mobility of electrons released by light in insulating photoconductors have used continuous illumination and have been steady-state dc measurements using an electrometer to measure the Hall voltage. These measurements have suffered from uncertainties due to buildup of space charge in the crystal near the current electrodes. Although corrections for such space charge can be made,[3] these corrections are of questionable accuracy. A technique which avoids these difficulties is described in detail in this paper. This technique has been applied to the alkali halides[4] and in modified form to silver chloride.[5] In this paper some experimental results on diamond are presented. Diamond was studied because it has a high electronic mobility and high photoconductive sensitivity, and because electronic mobility theory in diamond is well developed.

MacDonald and Robinson have recently used an alternative technique[6] for making such measurements which, where applicable, is more accurate, but which requires more light and/or greater photoconductive sensitivity. Unfortunately the author was not aware of their work until research for the present paper was almost complete. It appears likely that further improvements in these measurements can be attained through the use of combinations of some of the ideas of both methods.

B. EXPERIMENTAL METHOD

The electrode arrangement first used in this work is shown in Fig. 1. The crystal is sandwiched between plates of electrically conducting glass, that is, glass coated with a very thin transparent semiconducting film. The conducting plates and the batteries across them set up a fairly uniform field in the crystal in the x direction. The potential of the entire upper plate is held constant during the experiment by the electrometer circuit, while the potential of the entire lower plate can be varied by varying the potentiometer setting. Doing this makes opposite points on the two conducting plates have different potentials, and inintroduces a small y component of electric field in the crystal. Adding the y component of electric field to the large x component of field gives a field at an angle to the x axis; thus, moving the potentiometer rotates the electric field inside the crystal through a small angle about the z axis.

The electrometer responds only to charge flow in the y direction. A charge flow in the x direction will produce no deflection. The situation is analogous to that in an ionization chamber; the fact that the electrodes have a built in potential gradient does not alter the situation appreciably.

To make a measurement, the magnetic field is turned on in the z direction, and the crystal is illuminated with flashes of light a few seconds long. The potentiometer is adjusted until there is no charge flow registered by the electrometer upon illumination. This means that the charge flow $\mathbf{I}$ is in the x direction, as mentioned above, but due to the Hall effect the electric field is at an angle to the x direction [Fig. 2(a)]. The magnetic field is then reversed and the potentiometer readjusted until the electrometer again gives no deflection upon illumination of the crystal. The situation in Fig. 2(b) then obtains. The current is again in the x direction but the electric field has been rotated with the potentiometer through twice the Hall angle. The

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[1] H. Lenz, Physik. Z. 25, 434 (1924).

[2] J. Evans, Phys. Rev. 57, 47 (1940).

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[4] A. G. Redfield, Phys. Rev. 91, 753 (1953); following paper, Phys. Rev. 94, 537 (1954).

[5] F. C. Brown, Phys. Rev. 92, 502 (1953).

[6] J. Ross MacDonald and John E. Robinson, Phys. Rev. (to be published). The author is indebted to Dr. MacDonald for a discussion of this work.

Hall angle can be approximately obtained from the voltage change ΔV_T and the geometry of the electrodes:

$$\frac{2\mu_H H}{c} \simeq 2\theta \simeq \frac{\Delta E_y}{E_x} \simeq \frac{\Delta V_T/D}{V_L/L}, \tag{1}$$

where μ_H and θ are the Hall mobility and angle, H is the magnetic field, c is the velocity of light, D is the thickness of the crystal, L is the length of the resistance film and ΔE_y is the change in E_y due to changing the potentiometer voltage by ΔV_T. This equation holds for small θ. We will show later how it can be corrected for edge effects and other errors.

This procedure is repeated for other values of the magnetic field to see whether the Hall voltage varies linearly with the magnetic field.

To eliminate the effects of charge which would build up near the surfaces of the crystal, the potentiometer is actually adjusted during a measurement in the following way: The batteries are all turned off and a bright light is shone on the crystal, releasing charge and neutralizing any residual space charge in the crystal. The light is then turned off and the batteries turned on. The crystal is illuminated with a weak light and the *initial* deflection of the electrometer is observed. The potentiometer is adjusted accordingly, i.e., to reduce or reverse this initial deflection, and the batteries are turned off again. This cycle is repeated until the electrometer gives no deflection upon illumination. The charge observed on the electrometer is at most a few times 10^{-14} coulomb, and it can be shown that this much charge flow produces a negligible effect on the electric field inside the crystal.

If the crystal shows appreciable dark current, charge will flow as soon as the batteries are turned on. This current will make the experiment impossible because the electric field inside the crystal will be distorted and destroyed and because it will be impossible to separate the dark current from the photocurrent. Thermally delayed or secondary photocurrent may give trouble for the same reason. These difficulties can sometimes be avoided with various modifications,[5] but in this paper we will discuss only completely insulating photoconductors showing no delayed photocurrent.

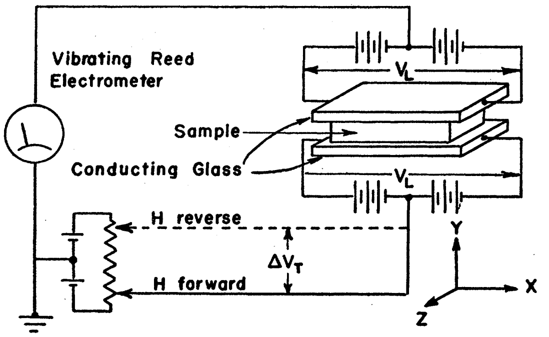

FIG. 1. Original electrode arrangement and circuit diagram.

This method differs from the conventional one in several interrelated ways. First, it is a transient method. Second, the electric field is rotated by external electrodes, rather than by the electrons under study. Third, the direction of charge flow is deduced from an electrometer measurement, rather than by confining the current to a thin sample whose orientation gives the direction of charge flow. Finally, no attempt is made to inject or eject electrons through the surface of the sample; the whole crystal takes part in the experiment. The method is similar in some respects to Freeman's method of measuring the Hall mobility of electrons released in diamond by beta particles.[7] The technique of using weak flashes of light to avoid space charge effects in photoconductivity experiments was originated many years ago by Pohl and his co-workers.[8]

The arrangement of Fig. 1 was actually used in some early measurements on diamond, but two serious difficulties arose. First, the upper plate had to be well insulated electrically from ground, and this meant that it was also well insulated thermally. This seriously limited the applied electric field because of the heat dissipated in the upper plate for high field values. Second, the upper battery had to be well insulated and shielded from ground, and the large size of the battery box resulted in unduly large noise and drift in the electrometer from ionization due to radioactive contamination.

The most important modification of the experiment is simply to remove the upper battery and connect the the electrometer directly to the upper plate (Fig. 3). The resulting electric field is highly nonuniform, but the principle of the experiment is exactly the same. The only difference is that for a given Hall angle and potential gradient on the resistance film the Hall voltage will only be about half as much, because the average x component of electric field is cut in half. The only modification of Eq. (5) is that E_x is replaced by $\bar{E}_x$, the average value of the x component of electric field in the crystal:

$$2\theta = 2\mu_H H/c = \Delta V_T / D\bar{E}_x, \tag{2}$$

where

$$\bar{E}_x = \int\int\int n_T E_x d^3\mathbf{x} \Big/ \int\int\int n_T d^3\mathbf{x}. \tag{3}$$

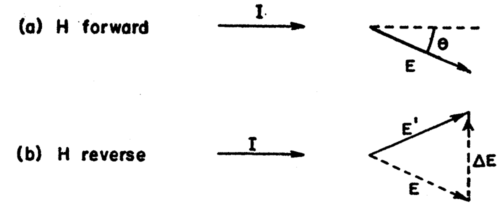

FIG. 2. Electric field and charge flow during the experiment.

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[7] G. P. Freeman, thesis, Leiden, 1952 (unpublished).

[8] N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1948), Chap. 4.

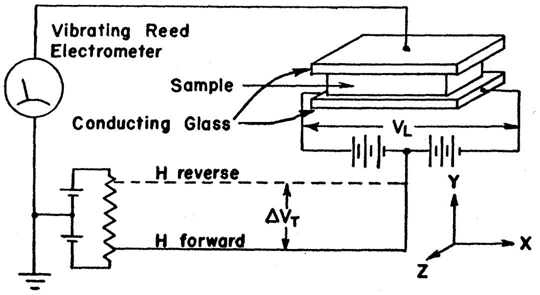

FIG. 3. Modified circuit diagram.

Here the integrations are over the volume of the crystal; n is the number of electrons per unit volume freed by the light pulse, and τ is the mean lifetime of the electrons before retrapping or annihilation. Equation (2) is proven in the appendix, and is valid for small values of θ, i.e., to first order in H. The only assumptions made in deriving Eq. (2) are that μ_H and μ_D, the Hall and drift mobilities, are constant throughout the crystal, that the area of the electrodes is much larger than the area of the sample, and that the edges of the sample are perpendicular to the electrodes.

Equation (2) applies to the arrangements of both Figs. 1 and 3. For Fig. 1, $\bar{E}_x \simeq V_L/L$ if the sample is thin; for Fig. 3, $\bar{E}_x \simeq \frac{1}{2} V_L/L$. For a thicker sample, $\bar{E}_x$ will be smaller due to the fringing field at the edge of the sample. Calculation of $\bar{E}_x$ is simply a problem in electrostatics which can be solved exactly in a few cases.

A disadvantage of the modification of Fig. 3 is that a slight amount of bleaching of the photoconductivity may make it impossible to get a reproducible setting of the potentiometer. This is because slight spatial inequalities in the amount of bleaching will cause the average position or centroid of the photoconductivity to move. With the magnetic field off, the proper potentiometer balance will be at the point where the y component of electric field is zero at the photoconductive centroid. If the centroid moves 1 percent of the length of the crystal, the proper potentiometer setting will move to change the voltage of the lower plate by 1 percent of the applied voltage V_L. This may be a large fraction of the Hall voltage. Similar arguments apply with the magnetic field turned on, and thus bleaching will give nonreproducibility and error in the measurement of ΔV_T, the Hall voltage. The percentage error so introduced can be reduced by making the crystal thicker, thus increasing the Hall voltage, but beyond a certain point this is disadvantageous because $\bar{E}_x$ is reduced.

For a given Hall angle and crystal thickness the measured Hall voltage, ΔV_T, is proportional to $\bar{E}_x$, and therefore it seems logical to try to make $\bar{E}_x$ as large as possible. E_x is obviously larger near the lower plate, a fact which suggests a further modification. If only part of the sample is photoconducting (Fig. 4), the parameter $n\tau$ is obviously zero for the upper part, and roughly constant for the lower part of the sample. If the photoconducting part is thin enough, $\bar{E}_x$ will almost equal V_L/L, rather than one-half V_L/L as in Fig. 3; if the photoconducting part is thick $\bar{E}_x$ will be somewhat less. As before, determination of $\bar{E}_x$ is simply a problem in electrostatics.

This modification has the advantage that, all other things being equal, the Hall voltage can be made as large as desired simply by increasing the thickness of the upper, nonphotoconducting part of the crystal. The required transverse field change in the photoconducting part remains constant because the Hall angle and $\bar{E}_x$ approximately do, but a larger applied voltage change ΔV_T is required to produce this given field change if the thickness is greater. This means that for samples of limited size, such as diamond, the Hall voltage can be increased several times, giving greater accuracy. Although the sensitivity of the electrometer for a given motion of charge is reduced in proportion to the thickness D, the noise due to fluctuations in the resistance film is similarly reduced, giving a constant signal-to-noise ratio. Furthermore, $\bar{E}_x$ is increased by a factor of approximately two, because the upper electrode no longer acts to reduce E_x in the photoconducting part of the sample, increasing ΔV_T by a factor of about $2D/\delta$. This modification also reduces bleaching errors by allowing the use of a thick crystal without reducing $\bar{E}_x$.

The main limitation of this method is that space charge difficulties are increased as D is made larger relative to δ, because a larger amount of charge flow is required to get a given accuracy.

Although a flat, thin sample is required for quantitative measurements, rough samples having a more or less flat shape can be used with the arrangements of either Fig. 3 or Fig. 4 if a crude measurement or a temperature dependence is all that is desired. Such measurements are useful in the case of diamond, because of the expense involved in grinding a diamond flat. Measurements can presumably also be made on polycrystalline, sintered, or powdered samples, although the possibility of space-charge difficulty is greater than with single crystals.

Further modifications of the method are the use of ac or pulsed electric and magnetic field, and chopped or very short pulsed light, with a narrow or wide band vacuum tube amplifier as a Hall detector. The advantages are reduction of noise error, and the possi-

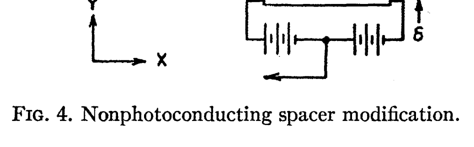

FIG. 4. Nonphotoconducting spacer modification.

bility of application to materials showing large delayed photocurrent or dark current. These variations were not used in this research, but Brown[5] has measured the Hall mobility of electrons in AgCl using a high speed pulsed light and pulsed electric field modification of the present method.

Evaluation of the parameter $\bar{E}_x$ defined in Eq. (3) is a straightforward boundary value problem if the sample is a circular cylinder or a rectangular parallelepiped with an infinite (or very large) z dimension. For a cylindrical sample in the arrangement of Fig. 4,

$$\bar{E}_x/E_0 = 1 - b/2 - \frac{2i(\epsilon-1)}{ab} \times \sum_{n=1}^{\infty} \frac{1-\cos(n\pi b)}{(n\pi)^3\left[\epsilon\frac{J_0(in\pi a)}{J_1(in\pi a)} - \frac{H_0^{(1)}(in\pi a)}{H_1^{(1)}(in\pi a)} - \frac{\epsilon-1}{in\pi a}\right]}, \quad (4)$$

where E_0 is the potential gradient on the lower plate, ϵ is the dielectric constant, a is the ratio of the radius of the cylinder to its thickness D, and b is the ratio of δ to D (Fig. 4). For a rectangular parallelepiped,

$$\bar{E}_x/E_0 = 1 - \frac{b}{2} - \frac{2(\epsilon-1)}{ab} \sum_{n=1}^{\infty} \frac{1-\cos(n\pi b)}{(n\pi)^3[1+\epsilon\coth(n\pi a)]}, \quad (5)$$

where a is one-half the ratio of the thickness in the x direction to that in the y direction, and the other symbols are the same. It turns out that $\bar{E}_x$ as given in formulas (4) and (5) is almost the same for a cylinder as for a rectangular parallelepiped, if the parameters a and b are, respectively, the same, so it seems reasonable to assume that $\bar{E}_x$ for the intermediate case of an elliptical slab is also almost the same function of a and b, with a equal to one-half the ratio of the maximum thickness in the x direction to that in the y direction. Most of the samples used in this experiment were reasonably close to elliptical slabs and this approximation was used to calculate $\bar{E}_x$.

If the charge sensitivity of the electrometer is too low or if there is a large amount of noise, the charge flow required to make a measurement may be so large that the electric field in the crystal will be appreciably distorted by space charge. In the work reported in this paper the charge flow registered by the electrometer was of the order of 10^{-14} coulomb per light pulse, and because of the internal consistency of the data it is believed that space-charge effects were negligible. This belief is confirmed by an analysis of the problem, the details of which we omit. This analysis rests on the fact that the electric field in the photoconducting sample after illumination is the same as that in a perfectly insulating sample having a higher dielectric constant, because both the photocurrent in a photoconductor and the dielectric polarization in an insulator are in the direction of the electric field and proportional to it (neglecting the Hall effect, which produces only a second-order error). This assumption is valid only for relatively small charge flow.

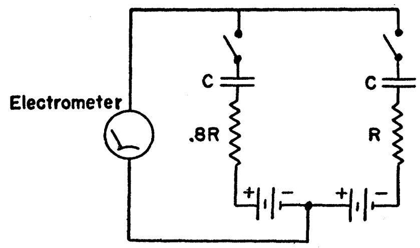

FIG. 5. Lumped circuit approximation of Fig. 4.

When the crystal is illuminated the electrometer behaves in a rather unexpected way. If the potentiometer is set at one side of the "true" setting corresponding to the applied magnetic field, the electrometer needle, which registers charge (not current), will move, say, in a positive direction, with a velocity which appears to increase with time. If the potentiometer setting is now moved to the other side of the "true" setting, and the accumulated space charge in the crystal destroyed by shining in a bright light with the batteries off, the electrometer needle will initially move in the negative direction upon illumination with the batteries on. But after a deflection of about 10^{-14} coulomb (in a typical case) the electrometer needle will slow down, stop, reverse, and move in the positive direction with increasing velocity. If the potentiometer is moved farther in the same direction, the electrometer excursion will be farther but the general behavior will be the same. This effect is surprising because one generally expects an exponentially decaying current due to the buildup of space charge. The effect occurs with or without a magnetic field, and is most pronounced when only part of the sample is photoconducting.

The most likely cause for this effect can be explained with the aid of Fig. 5, which is supposed to approximate the arrangement of Fig. 4. The two resistors represent the two halves of the photoconducting part of the crystal, and the two condensers the two halves of the upper part of the crystal. Turning on the switches is equivalent to turning on the light, and the fact that the two resistors are unequal is supposed to represent the fact that the crystal may not be uniformly illuminated. When the switches are turned on, the condensers are uncharged and the current flow in the left-hand arm is opposite to and larger than that in the right-hand arm, so the net charge flow is clockwise. However, the left-hand condenser will charge up faster, the current in the left-hand arm will become smaller, and after a certain time the current will decrease, stop, and reverse, and the electrometer will eventually return to its initial reading. The latter fact is due to the equality of the condensers and the equality and opposite polarity of the batteries.

This simple analog shows that the current reverse effect is due to unequal polarization and illumination in different regions of the crystal. It still appears, however, that this effect will give no error as long as the initial deflection of the electrometer is used in making observations. The charge value at which the current reverses is about what one would expect from space charge considerations.

The most serious noise in this experiment was caused by the light itself. This manifested itself in a swing of the electrometer needle in one direction when the light was turned on, and a swing in the reverse direction when the light was turned off. If the "sample" was not a photoconductor, the needle would return to the same reading, plus or minus a small amount due to drift. The time required for the electrometer to reach an equilibrium reading when the light was turned on or off was about 10 seconds.

This spurious signal could be divided into two parts: The first was due to photoconductivity in the resistance film, which will give a signal because the illumination and the resulting resistance change of the film may not be uniform, and a nonuniform change of resistance will result in a change in the average potential of the lower plate. This potential change will be capacitatively coupled to the electrometer. Only certain kinds of film show this photoconductivity.

The other part of the spurious signal occurred even when the batteries were turned off, so that film photoconductivity could not be responsible. It seems likely that this signal was due to the heating of some part of the crystal holder or the crystal, because it occurred most strongly at low temperatures (80°K) where the thermal contact between different parts of the system was poor; however, the signal was not due to thermal expansion of the sample, as could be proven by applying a large voltage V_T to the lower plate, with V_L off, and observing that the signal was unchanged. The exact origin of this signal is still somewhat of a mystery.

Another serious kind of noise originating in the resistance film occurred when the film was carrying current and was apparently similar to the noise which occurs in carbon resistors. This noise was serious only in certain types of film.

If the sample used is not uniformly photoconducting due to inhomogenieties in the impurity content or to absorption of light, $\bar{E}_x$ may be different from the calculated value. Since E_x varies from E_0 to zero between the bottom and top of the crystal this error may be quite important. If the error is due to inhomogeniety in the material, it can be greatly reduced by making a Hall measurement, turning the sample over, making another measurement, and averaging. A value obtained in this way is the same as that which would be obtained from the arrangement in Fig. 1, by the superposition theorem. The arrangement of Fig. 1 is not so susceptible to such errors because E_x is much more uniform throughout the crystal. If the error is due to light absorption, it may be calculable,[5] or it can be reduced by using a crystal of dimensions large compared to the light absorption length, and illuminating through the bottom plate. In this case $\bar{E}_x$ will be very nearly equal to E_0.

Other possible kinds of noise and error which were unimportant in practice are mechanical vibration, battery noise and drift, electrometer and insulator noise, surface leakage, Hall effect in the resistance film, and thermoelectric effects. The voltage applied to the resistance film, and thus $\bar{E}_x$, was limited by heating effects in the film, and the resulting noise and drift in the electrometer.

C. EXPERIMENTAL APPARATUS

The cryostat is diagramed in Fig. 6. This cryostat cools the crystal by conduction through a heavy copper bar which is in contact with an external temperature bath in a Dewar flask. The tail of the cryostat was immersed in a coolant which was ice water, dry ice and acetone, or liquid nitrogen.

The cryostat was evacuated with a mechanical pump and the pressures attained were of the order of a few microns. The vacuum served both to insulate the crystal holder and heat conductor thermally, and to reduce surface leakage current and radioactive ionization noise in the electrometer.

The copper heat conductor was in two sections, held together with a $\frac{1}{4}$-20 stud bolt and soldered with Wood's metal for effective heat conduction and ease of removal. The temperature drop between the coolant and the base

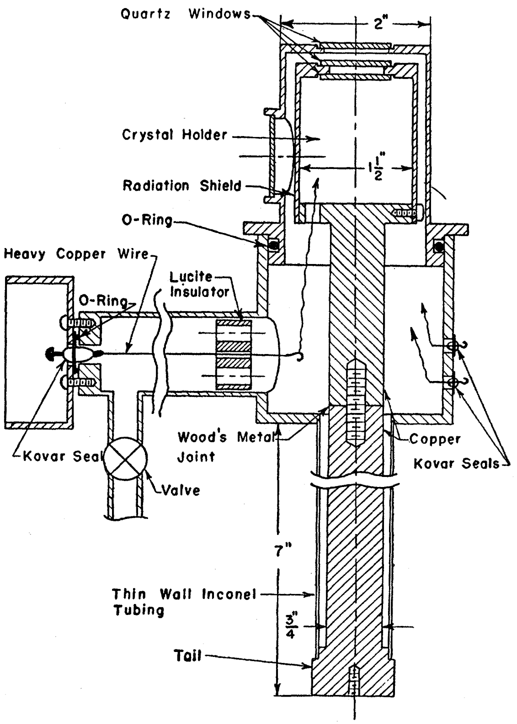

FIG. 6. Simplified cross section of the cryostat.

of the crystal holder was normally about four degrees at 80°K.

The cryostat was held below the gap of the magnet in such a way that the crystal holder and the two-inch diameter vacuum jacket around it projected up into the bottom part of the magnet gap. In this way it was possible to fit into a magnet which was already occupied with another experiment and which would otherwise have been unavailable for this work.

The lead from the crystal holder to the electrometer head consisted of a number 30 constantan wire leading to a heavy copper wire, which went down the center of a long (two-foot) tube to a Kovar seal. Constantan wire was used to minimize heat conduction. The only insulators used between the crystal holder and the electrometer were the Kovar seal and a piece of Lucite, both of which could be removed for cleaning. The Kovar seal was vacuum sealed to the end of the tube by being clamped down hard against a greased O ring. The assembly which clamped the O ring was also designed to fit the head of the vibrating reed electrometer,[9] and the sensitive lead of the electrometer made direct contact with the knob on the end of the Kovar seal. The same pipe which was used to lead out the electrometer connection was also the pumping lead, and the Lucite insulator had several holes drilled through it to reduce its pumping impedance.

Temperatures between ice, dry ice, and liquid nitrogen temperatures were obtained in one of two ways. The first was to cool the cryostat to low temperature with liquid nitrogen and keep the Dewar flask in position, but then remove enough liquid nitrogen so that the tail of the cryostat was no longer in contact with it. Doing this minimized warming of the copper rod by air convection, and the crystal holder warmed up slowly (from 100 to 200 degrees kelvin in about one hour) due to conduction up the inconel tube and radiation.

In the other method for attaining intermediate temperatures a copper appendage was screwed to the end of the tail with a spacer of brass or copper, and the appendage was placed in contact with the temperature bath. After a long time the copper bar would come to equilibrium at a temperature somewhat above that of the coolant. Although this arrangement gave a more stable temperature, it was rather unsatisfactory because the temperature attained depended strongly on the nature of the contact between the spacer and the two pieces of copper, and it was impossible to predict or prearrange the temperature at which the cryostat would reach equilibrium.

Compared to the more usual optical cryostat design, in which the crystal holder is attached to the bottom of the coolant holder of a Dewar-shaped vessel, this design has the advantages of simpler construction, ease of quick warming, and (what was most important in the present work) the possibility of being placed unobtrusively in the gap of a busy magnet. Its disadvantages are high liquid nitrogen consumption and slowness in cooling.

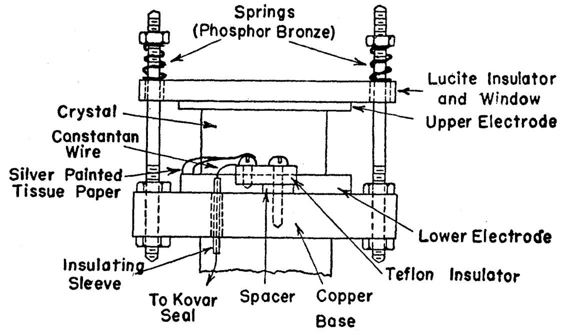

FIG. 7. The crystal holder. The radiation shield, which is omitted, fits over the whole assembly as indicated in Fig. 6. The thermocouples and two of the three electrical connections to the electrodes are also omitted.

About six different crystal holders were tried during the experiment, and the trouble with most of the early models was that the crystal was not sufficiently well shielded from thermal radiation from the outside world. The solution was to surround the crystal completely with a radiation shield held at the temperature of the crystal. A hole in the shield as small as one-quarter inch in diameter a centimeter or so from the crystal could easily raise its temperature by several degrees, which is a serious error.

A fairly satisfactory crystal holder design is shown in Figs. 6 and 7. The radiation shield, shown in Fig. 6, was screwed to the circular base with six radial screws. The two quartz windows were clamped to the top of the shield with stiff phosphor bronze springs (not shown) and the surface where they made contact with the copper shield was greased with silicone vacuum grease for better thermal contact. Silicone grease was used because of its low hardening temperature. A quartz window makes a good radiation shield because it absorbs strongly in the far infrared and thus absorbs thermal radiations from the outside, re-emitting thermal radiation characteristic of its own temperature. The outer window of the radiation shield was supposed to act as a radiation shield for the inner window, so that despite the probable poor thermal contact between the windows and the shield, the temperature of the inner window was probably very close to that of the rest of the shield.

The essential features of the crystal holder with the radiation shield removed are shown in Fig. 7. The lower electrode was insulated from the base with one or two 0.001 inch sheets of du Pont Mylar plastic film, which has great mechanical and dielectric strength. In the case of diamond, a sheet of Mylar was also placed between the crystal and the lower electrode to prevent scratching of the resistance film. Silicone grease was used liberally at all interfaces between the crystal, the electrically conducting glass, the base, and the various

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[9] Manufactured by the Applied Physics Corporation, Pasadena, California.

pieces of Mylar film. The purpose was to make the thermal contact between the crystal and the base as good as possible. The grease was effective in this respect down to about 180°K, but below this temperature the grease solidified and cracked, because of differential thermal expansion of the various parts of the holder. It was below this temperature that good radiation shielding was really important.

Two different types of conducting glass were used. One was a Nesa[10] coating which had a room temperature surface resistivity of about 200 000 ohms (per square), and surface resistivity of about 60 megohms at 80°K.

The principle trouble with this film was that its low resistance limited the electric field because of heat dissipation problems to 400 volts per centimeter or less. At low temperature it also showed very serious photoconductivity.

The other conducting glass used was very kindly supplied by Dr. K. B. Blodgett of the General Electric Research Laboratory, and was made by heating lead glass at high temperature (about 500°C) in a stream of hydrogen.[11] This film had a room temperature surface resistivity of about 5000 megohms, and a surface resistivity at 80°K of about 100 000 megohms. This film did not show any photoconductivity, but gave serious noise whenever a voltage was applied to it. The noise was less at low temperatures, and a few measurements were made at 80°K using this film. Its higher resistance allowed the use of higher electrical fields in the crystal.

The linearity of the potential gradient on the surface of the film could be checked with a probe connected to a high-resistance potentiometer. Such measurements were made at room temperature, but it seems reasonable to assume that a film which is uniform at room temperature will also be uniform at lower temperatures.

The upper electrode was either a piece of Nesa-coated glass or a plain piece of glass painted with closely spaced stripes of silver paint.

An important use of the resistance films was to act as a check on the temperature of the crystal. If the temperature of the lower resistance film was the same as that of the base of the crystal holder, it was assumed that the radiation shielding was adequate and that the crystal was also at the same temperature. On the other hand, if the resistance film temperature was above that of the base the crystal temperature would certainly be higher also.

The temperature of the resistance film could be inferred from measurements of its resistance. The resistance was reliably known from 200 to 300°K, where the silicone grease could be relied upon to make good thermal contact. The resistance at 77°K was known from measurements made while the film was immersed in liquid nitrogen. The resistance at intermediate temperatures could not be measured directly, but the effects of inadequate radiation shielding could easily be detected because the film resistance was not a reproducible function of temperature (as measured with a thermocouple screwed to the base), and the resistance did not approach the known value at 77°K. With the crystal holder of Figs. 6 and 7 the resistance-temperature function was reasonably smooth, although there was still some scatter at the lowest temperatures. This scatter may have been due to very long time constant photoconductivity (for which there is some evidence) in the case of the Nesa film, and to inaccuracies in the resistance measurement in the case of the lead glass film.

The resistance measurements indicate that for the crystal holder of Figs. 6 and 7 the crystal temperature was probably less than four degrees below the temperature of the base. There is, however, a slim possibility that the difference was as much as 8 degrees.

In other crystal holder designs the plane of the electrodes was vertical and the illumination was horizontal through the window in the side of the vacuum jacket. Such an arrangement has the disadvantage that less space is available for the crystal; on the other hand, the crystal can be illuminated through the "bottom" electrode, i.e., the electrode connected to the batteries V_L, through a hole drilled the base of the holder. This is desirable if the crystal used absorbs light strongly, but it was not particularly advantageous in the present research.

The magnet was designed and built for nuclear resonance work by H. W. Knoebel, R. E. Norberg, and D. H. Holcomb, and was very kindly made available for this research by Professor C. P. Slichter. The magnetic field could be varied from zero to ten kilogauss. The crystal was an inch or more from the edge of the magnet gap, and flip coil measurements showed that the field at this point was 1 percent $\pm\frac{1}{2}$ percent less than the field at the center of the gap.

The optical system was exceedingly crude, and consisted of a 250-watt 110-volt sealed beam spotlight and a collection of filters. The lamp was supplied through a variac from the 110-volt ac line, and could be turned on with a pushbutton switch on the output of the variac. To eliminate electrical interference in the electrometer a Cornell-Dubilier type IF-18 interference filter was connected between the variac and the switch. A glass absorption cell containing a few percent solution of cupric chloride in distilled water was used to remove infrared radiation from the lamp. The spotlight was roughly $1\frac{1}{2}$ feet from the crystal. It was run at between 40 and 110 volts for the observational pulse of light, and usually at higher voltage for the removal of space charge.

All voltages were supplied by batteries. V_L was between 90 and 720 volts, depending on the tempera-

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[10] Trade name for electrically conducting glass manufactured by the Pittsburgh Plate Glass Company. See L. B. Clark, Sr., Rev. Sci. Instr. 24, 641 (1953); R. C. Gomer, Rev. Sci. Instr. 24, 993 (1953).

[11] K. B. Blodgett, J. Am. Ceram. Soc. 34, 14 (1951).

TABLE I. Summary of the characteristics of all the diamonds studied.

Diamond	uv transmission at 2537A	Fluorescence	Mobility cm²/volt sec		Thickness mm	½ length / thickness	½ width / thickness	$\bar{E}_x/E_0$ assumed
			300°K	200°K				
A	No	Yes	−720	−1300	1.2	1.45	0.9	0.435
PS	No	Very small	−1850	−3350	1.3	1.8	1.8	0.45
US1	Yes	Very small	−1700	−3150	0.35	4.5	2.7	0.48
US2	No	Very small	(−900)	(−1200)				
US3	Yes	Very small	Low photocond.					
US4	No	Very small	(−900)	(−1000)				
US5	No	Very small		(−1200)				
US6	No	Very small	Not investigated					
R1	No	Very small	(+1100)	(+1400)				
R2	No	Very small	(−800)	(−1000)				
R3	Yes	Very small	−1400	−2250	0.78	3.0	1.63	0.47
R4	No	Very small	Dark conductivity					
R5	No	Very small	Dark conductivity					
R6	No	Very small	−1200	−1700	0.66	2.6	1.8	0.465
R7	Yes	Very small	Dark conductivity					
R8	Yes	Very small	Dark conductivity					
R9	Yes	Very small	Dark conductivity					
R10	Yes	Very small	+1000	+1400	1.07	1.42	1.0	0.435
R11	Yes	Very small	+1200	+2200	0.975	1.54	1.24	0.44
R12–17	No	Very small	Not investigated					
R18–20	No	Yes	Not investigated					

ture and resistance film, and the Hall voltage ΔV_T was as much as 90 volts. At the higher temperatures it was necessary to use a lower V_L because the film resistance was lower, and therefore, the accuracy was lower.

D. EXPERIMENTAL RESULTS

The vital statistics for the diamonds studied are given in Table I, together with some rough data on the magnitude and temperature dependence of the mobility.

The samples R1 to R20 were purchased from the Rough Diamond Company, New York, New York; and samples US1 to US6 were purchased from the U. S. Industrial Diamond Company, New York, New York. Some of these were apparently whole rough diamonds and others were cleavage plates, but all were thin plates with more or less flat surfaces.

Diamond PS was the same sample studied by Pearlstein and Sutton in their measurements of the electronic drift mobility.[12] Diamond A had been previously studied by Raboy[13] and was known not to count alpha particles.

The luminescence was investigated with light from a low-pressure mercury arc filtered with a Corning No. 7–54 filter. The luminescence was blue or blue-green as reported by other investigators. The ultraviolet transmission was investigated using the same source and filter, and detecting the radiation with willemite powder. A previous crude experiment had shown that this powder was sensitive mainly to radiations below 3000A, so that this test was essentially a measure of the optical transmission of the diamond at 2537A. The results of this test were verified in the case of diamonds R3, US1, and PS with transmission spectra taken with a quartz spectrograph and a hydrogen arc.

As indicated in Table I, many of the diamonds did not photoconduct sufficiently, or showed dark conductivity. In several cases this conductivity was apparently surface leakage, and could be eliminated by cleaning the diamond in hot nitric acid, cleaning solution, or aqua regia. Another effective cleaning procedure was to heat the diamond to orange heat with a torch, burning off a microscopic surface layer. This must be done cautiously, because diamond slowly changes to graphite at high temperatures. Several diamonds were treated in this way with no apparent ill effect.

Preliminary measurements were made on a number of rough diamonds. The method of Fig. 4 was used for greater relative accuracy. Silicone grease was used to fill the spaces between the crystal and the lower electrode caused by departures from flatness of the diamond surface. The purpose was to couple the crystal to the lower electrode electrostatically so that $\bar{E}_x$ would be as close to E_0 as possible. In some cases what corresponds to the "nonphotoconducting" part of the crystal in Fig. 4 was a 3.85-mm Teflon spacer. In these cases the diamond was illuminated through the "bottom" plate, as described in the last section. In other cases this spacer was a transparent insulator and the illumination was through the top. These measurements gave an accurate idea of the temperature dependence of the mobility, though because of the irregular shape of the diamonds it was possible to get only a rough idea of the absolute magnitude.[14] These measurements were

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[12] E. Pearlstein and R. B. Sutton, Phys. Rev. **79**, 907 (1950).

[13] S. Raboy, thesis, Carnegie Institute of Technology, 1950 (unpublished).

[14] To do this, the dielectric constant of diamond must be known. The value 16.5 quoted in various reference books is wrong; the correct value is 5.67. See S. Whitehead and W. Hackett, Proc. Roy. Soc. (London) **A51**, 173 (1939).

made with the tungsten lamp and the cupric chloride filter plus another blue filter with a gradual cutoff at about 5500A. In some cases a blue filter with a 4300A sharp cutoff was used. In no case did these filters make a significant difference in the measurement.

On the basis of this preliminary survey 7 interesting diamonds were chosen for further study, and their surfaces were ground and polished flat. This work was done by the Arthur A. Crafts Company, Chicago, Illinois. Only the top and bottom surfaces were ground flat; the edges were left rough. These seven diamonds were rerun after polishing using the method of Fig. 4 with a Teflon or sodium chloride spacer. As before, silicone grease was used to improve the electrostatic and thermal coupling between the crystal and the lower electrode. To get temperatures intermediate between liquid air, dry ice, and ice temperatures the cryostat was allowed to warm up slowly as described previously, and a series of measurements were made with the magnetic field alternately forward and reverse.

To get a more accurate value of the absolute magnitude of the mobility, measurements were made at dry ice and liquid air temperatures using no dielectric spacer above the crystal, as in Fig. 3. The relative measurements obtained as described in the previous paragraph were then normalized to agree with this absolute data. In Fig. 8 is plotted data for one of the most studied diamonds. The absolute measurements are denoted by the crosses, and the normalized relative data by elipses. The estimated maximum error is denoted by the size of the crosses and elipses. The measurements shown are the result of four different runs, two absolute and two relative.

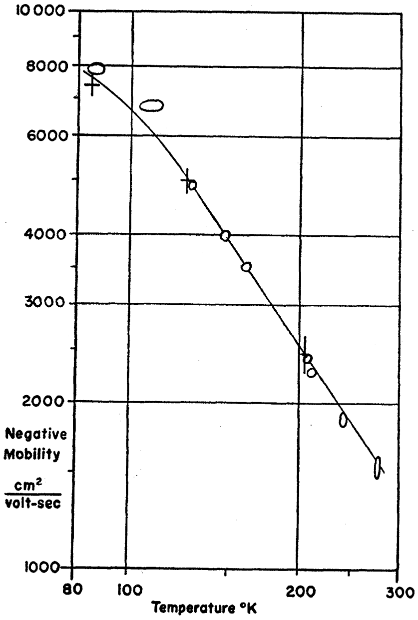

FIG. 8. Experimental data for diamond R3.

To reduce the effects of inhomogeneities in the diamonds, each absolute measurement was the average of two absolute measurements, with the diamond turned over between measurements. The ratio of the two measurements is a measure of the homogeneity of the diamond; if the ratio is one, the diamond is presumably fairly homogeneous. The observed ratios were always nearly the same at dry ice and liquid air temperatures, as expected. US1 and R3 consistently gave ratios of about 1.1:1 to 1.3:1; R6, R10, R11, and A gave ratios between 1.5:1 and 2.5:1; and PS was the least homogeneous, yielding ratios as high as 4:1. The ratio in PS was also not reproducible; a later measurement yielded a ratio of 2:1.

At temperatures near 100°K, where the Hall angle at 10 000 gauss approaches one radian in some samples, the apparent Hall mobility decreased as the magnetic field increased. That is, the ratio of ΔV_T to $\Delta H = 2H$ would decrease as H was increased. This decrease is attributed to third-order terms in H (second-order Hall effect). At these temperatures the measured mobility approached a value 15 ± 5 percent larger than the 10 000 gauss value, as H approached zero. This behavior was observed unambiguously only at low temperatures in the three high-mobility samples (R3, US1, and PS). It was observed in both the absolute and relative measurements on these samples. The true Hall mobility was assumed to be the mobility as H approaches zero. The departure from this value appeared to be proportional to H^2 as expected.

An attempt was made to investigate the spectral variation of the apparent mobility in diamond R3. All that was established was that at 100°K the measured mobility was smaller by about 10 percent at 5000A, as compared to 3650A. Above 5000A the mobility was of the same order of magnitude as below.

In all the measurements described above, the Hall voltage was proportional to the electrical field. That is, for any given magnetic field, V_T was always proportional to V_L.

The Hall voltage did *not* always vary linearly with magnetic field. A survey of all the measurements made on diamond indicates that there was no clear-cut correlation between the direction of the Hall voltage asymmetry and the orientation of the crystal holder, the electric field, or the magnetic field. For any given diamond in a given run the relative asymmetry was independent of the electric field, and reversed sign with the electric field, consistent with the previous paragraph. The only cases which showed a clear-cut asymmetry above error were those where an absolute measurement was being made with no dielectric spacer between the diamond and the upper electrode; i.e., where the electric field was most nonuniform. R3 showed far and away the greatest asymmetries; in some cases the asymmetry $[V_T(+H)+V_T(-H)-2V_T(0)]/2[V_T(+H)-V_T(-H)]$ was as much as 0.1 for $H=$

±10 000 gauss at 80°K. At temperatures above 200°K the asymmetry was negligible.

These asymmetries probably did not affect the Hall measurements, since these measurements were made by observing the required potentiometer voltage change when the magnetic field was reversed, rather than merely turned on or off. The origin of the asymmetry is probably magnetoresistance effect; arguments which will be omitted here indicate that the anisotropic magnetoresistance effect is the most important. This explanation is, however, by no means certain.

The dark resistivity of the diamonds on which successful measurements were made was apparently 10^{16} ohm-cm or greater at room temperature and below.

The mobility data for the most thoroughly studied diamonds is summarized in Fig. 9. The accuracy for most of the data is ±5 percent to ±10 percent. The discrepancy between R3, US1, and PS is apparently real, but due to some scatter of the data this cannot be said with certainty.

E. DISCUSSION

The data is consistent with previous Hall mobility measurements on diamond.[1,3,7]

The fact that three of the diamonds show a Hall effect characteristic of holes is in itself interesting, and indicates that both electrons and holes can be expected simultaneously in all the diamonds studied. It should be noted that the wavelength of the light used (about 4000A) was such that the electrons and holes could be excited from energy levels roughly in the center of the energy band gap, and it is perfectly plausible that such light could yield both electrons and holes simultaneously.

It seems certain that the temperature dependence of the observed mobility is determined in part by the temperature variation of the relative number of electrons and holes produced by the light. This makes interpretation of the data difficult.

In a very thorough study of the counting properties of diamond, Freeman[6,15] has shown that the best counting diamonds tend to be the most perfect ones. For this reason it is unlikely that, as has been sometimes suggested, mosaic or grain boundary conduction is important in the conduction counting mechanism. For the same reason it is believed that such processes are unimportant in the present research.

It is assumed that the diamonds used were perfect insulators throughout their volume. It is conceivable that the diamonds contained impure semiconducting domains which would have distorted the electric field. If this were the case, however, erratic noise and drift might be expected in the electrometer. No such behavior was normally observed below room temperature.

The most striking feature of the data is that the three diamonds PS, US1, and R3, which seem to follow a $T^{-\frac{3}{2}}$ temperature dependence (as predicted by the deformation potential theory)[16,17] also have the highest mobility, and differ in absolute magnitude by a relatively small amount. It seems likely that these diamonds give the true order of magnitude of the intrinsic electronic mobility. If there were a large number of holes in these diamonds it seems improbable that the measured mobility would follow the $T^{-\frac{3}{2}}$ law. To do this, the electron-hole density ratio would have to remain constant with temperature (assuming that the true electron and hole mobilities follow the $T^{-\frac{3}{2}}$ law).

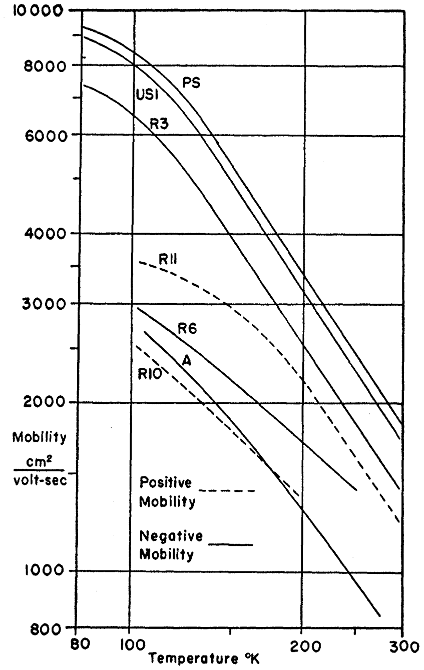

FIG. 9. Summary of the experimental data for the most thoroughly studied diamonds.

The difficulty with this interpretation is the 30 percent discrepancy between these three diamonds. This may conceivably be an experimental error. If the discrepancy is real it is probably due to the presence of holes in R3. If the electron-hole concentration ratio in R3 were constant to about 50 percent over the temperature range studied this would be a possible explanation.

We will tentatively assume that the electronic mobility is 1800 at 300°K. It seems certain that this is the correct order of magnitude, but on the basis of the existing data, an error of twenty to thirty percent is not inconceivable.

The measurements on R11 approach a $T^{-\frac{3}{2}}$ temperature dependence at high temperatures, and are characteristic of hole conduction. These measurements set a lower limit on the hole mobility of 1200 cm²/volt second at 300°K. Considering how close R11 approaches

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[15] G. P. Freeman and H. A. van der Velden, Physica **16**, 486 (1950); **17**, 565 (1951); **18** (1952).

[16] F. Seitz, Phys. Rev. **73**, 549 (1948).

[17] J. Bardeen and W. Shockley, Phys. Rev. **80**, 72 (1950).

the $T^{-\frac{3}{2}}$ dependence, it seems likely that the true hole mobility is not much larger.

The slight bend in the experimental curves of PS, US1, and R3 at low temperatures may be due to experimental error or impurity scattering. The similar shape of these curves suggests, on the other hand, that the bend may represent the true dependence of the mobility with only thermal scattering important. The temperature dependence of the remaining samples is undoubtedly determined by dislocation and impurity scattering, and by the presence of both electrons and holes.

The measured Hall mobilities are considerably lower than the drift mobilities observed by Pearlstein and Sutton[12] in diamond PS. It may be that the difference is caused by nonspherical energy surfaces in k space,[18] or nonisotropic thermal scattering of the electrons by the lattice. The discrepancy is, however, rather large for this to be the case.

The mobilities observed in the present research are consistent with the deformation potential theory[16,17] if the effective mass ratio is assumed equal to one and the deformation-energy band edge coefficients defined by Bardeen and Shockley[17] are taken to be $|E_{1c}|=7$ ev and $|E_{1v}|=8$ ev. These values are reasonable when compared with those for silicon and germanium, although direct application of the theory to some of these cases may not be possible, owing to probable complications in the band structure or the scattering dependence on velocity.[19] On the other hand, the drift mobility measurements of Pearlstein and Sutton[12] indicate that $|E_{1c}|$ and $|E_{1v}|$ may be smaller, or that the theory may also be inapplicable to diamond. In any case, it is clear that the relatively high value of the mobility in diamond is a partial consequence of its relative inelasticity, or, equivalently, its high Debye temperature.

Further investigation of the Hall effect in a large number of diamonds might yield a better picture of the photoconductive processes in diamond and less ambiguous values of the electron and hole mobilities. Variation of the Hall mobility at high electric fields should be an interesting extension of the investigation. Finally, study of the magnetoresistance effects in diamond should be possible using similar methods, and should give information about the energy band structure.

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F. APPENDIX

We now derive Eq. (2) above.

If an electron is freed in the crystal at a point $\mathbf{x}$, and moves a small distance $\mathbf{d}$ before retrapping or annihilation, the resulting charge flow registered by the electrometer will be

$$Q=e\mathbf{d}\cdot\partial\mathbf{E}/\partial V_T, \tag{6}$$

where e is the electronic charge, and V_T is the potentiometer voltage, and $\mathbf{E}$ is evaluated at the point $\mathbf{x}$. This result is independent of the electrode and sample shape and of the value or presence of the batteries V_L, and was proven for the vacuum case by Shockley;[20] a more general proof applicable to the present case follows from the Carson reciprocity theorem.[21]

If $n(\mathbf{x})$ electrons per unit volume are released in the sample, Eq. (6) becomes

$$Q=\int\int\int ne\mathbf{d}\cdot(\partial\mathbf{E}/\partial V_T)d^3\mathbf{x}. \tag{7}$$

The distance $\mathbf{d}$ traveled by the electrons (*shubweg*) is given by

$$\mathbf{d}=\mathbf{v}\tau=\tau\mu_D\mathbf{E}+\tau\mu_D\mu_H\mathbf{E}\times\mathbf{H}/c, \tag{8}$$

plus higher-order terms in $\mathbf{E}$ and $\mathbf{H}$. Here $\mathbf{v}$ is the electronic velocity, and τ is the mean time before trapping or annihilation of the released electrons.

In making a measurement, V_T is adjusted until Q equals zero. Using Eqs. (7) and (8) we get

$$Q=0=\int\int\int ne\tau[\mu_D\mathbf{E}\cdot(\partial\mathbf{E}/\partial V_T) + (\mu_D\mu_H/c)(\mathbf{E}\times H)\cdot(\partial\mathbf{E}/\partial V_T)]d^3\mathbf{x}, \tag{9}$$

plus higher-order terms.

Reversing the magnetic field and readjusting V_T an amount ΔV_T until $Q=0$ once again, gives

$$Q=0=\int\int\int ne\tau[\mu_D(\mathbf{E}+\Delta V_T\partial\mathbf{E}/\partial V_T)(\partial\mathbf{E}/\partial V_T) - (\mu_D\mu_H/c)(\mathbf{E}+\Delta V_T\partial\mathbf{E}/\partial V_T)(\mathbf{H}\times\partial\mathbf{E}/\partial V_T)]d^3\mathbf{x}, \tag{10}$$

plus higher-order terms.

Subtracting Eq. (9) from (10), and using the vector identity

$$\mathbf{A}\cdot(\mathbf{B}\times\mathbf{A})\equiv 0$$

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[18] W. Shockley, *Electrons and Holes in Semiconductors* (D. Van Nostrand Company, Inc., New York, 1950), p. 338.

[19] M. B. Prince, Phys. Rev. **92**, 681 (1953).

[20] W. Shockley, J. Appl. Phys. **9**, 635 (1938).

[21] J. R. Carson, Bell System Tech. J. **3**, 393 (1924); **9**, 325 (1930).

gives

$$\Delta V_T\int\int\int ne\mu_{DT}(\partial\mathbf{E}/\partial V_T)^2 d^3\mathbf{x}$$
$$=2/c\int\int\int ne\mu_D\mu_{HT}(\mathbf{E}\times\mathbf{H})\cdot\partial\mathbf{E}/\partial V_T d^3\mathbf{x}. \quad (11)$$

This equation is exact to second order in **H**. The second-order (magnetoresistance) terms drop out because the measurement is made by reversing the magnetic field, not simply turning it on and off.

Equation (11) clearly applies to the arrangements of Figs. 1, 3, or 4. In these cases we can assume that $\partial\mathbf{E}/\partial V_T$ is a vector in the y direction of magnitude $1/D$, because Laplace's equation and the boundary conditions will then be satisfied. We can also assume that μ_D and μ_H are constant, and that **H** is uniform in the z direction. Under these assumptions Eq. (2) follows immediately.

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Electronic Hall Effect in the Alkali Halides*

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The Hall mobility of electrons in colored NaCl, KCl, KBr, and KI has been measured using an unconventional technique previously described by the author. Measurements on additively and x-ray colored NaCl yield an electronic mobility of 250±50 cm²/volt sec at 84°K, decreasing to about 25±25 at 200°K. In additively colored KCl the mobility is 100±50 at 90°K; in additively colored KBr it is about 110 at 84°K. In additively colored KI the mobility is 155±30 at 84°K, decreasing to 30±15 at 200°K. In every case the sign of the Hall effect is electronic. These observations are consistent with the mobility theory of Low and Pines assuming effective mass ratios between 0.35 and 0.6 for the unpolarizable crystal, or polaron effective mass ratios between 0.5 and 0.9. These mobility data combined with photoconductive yield data give values of the capture cross section of F centers for electrons (to form F' centers) between 0.7 and 5 times 10^{-15} cm² at about 200°K.

A. INTRODUCTION

THIS paper reports some measurements of the electronic Hall mobility in colored NaCl, KBr, and KI using techniques described in the immediately preceding paper[1] (henceforth referred to as EHED). Previous investigations[2–4] in these crystals failed to give quantitative values of the mobility, although Evans[4] established an upper limit of 20 cm²/volt sec at 300°K for the mobility in several colored alkali halides. While the present work was in progress, MacDonald and Robinson[5] succeeded in measuring the electronic mobility in KBr, obtaining a value of 12.4 cm²/volt sec at 300°K.

This work was undertaken mainly to test the theory of electronic mobility in polar substances.[6–8] Theory predicts that the mobility will vary with temperature as $\exp(\Theta/T)$, where Θ is the Debye temperature corresponding to the longitudinal optical modes of vibration of the lattice, and provided $T\ll\Theta$. Θ can be deduced[9] from the infrared absorbtion peak[10] of the crystal and its low- and high-frequency dielectric constants.[11] The values of Θ for the crystals investigated in this research are NaCl, 370°K; KCl, 300°K; KBr, 236°K; KI, 190°K. Thus the theory applies to those alkali halides only considerably below room temperature.

The electrons in this experiment are obtained by ionizing F centers with light of the proper wavelength. The F centers act only as sources of electrons, and in view of their small concentration they should not affect the experimental measurements.

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[1] A. Redfield, preceding paper, Phys. Rev. **94**, 526 (1954).

[2] P. Lukirski, *Reports of the Physical-Technical Roentgenological Institute and the Leningrad Physical-Technical Laboratory 1918–26* (Leningrad, 1926), p. 185.

[3] P. Tartakowski, Z. Physik **66**, 830 (1930).

[4] J. Evans, Phys. Rev. **57**, 47 (1940).

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