

Temperature Dependence of Electron Mobility in AgCl†

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By using improved crystal counter techniques, electron mobility has been investigated as a function of temperature in AgCl. The measurements were made on annealed samples carefully grown from the melt in which the range of conduction electrons was of the order of 10^{-4} cm per volt/cm of electric field strength. At high fields electron trapping in the volume of the crystal is less important and observed mobility is shown to become a constant independent of field. At 86°K, drift mobility was found to be 274 cm²/volt sec and was reproducible to within 10 percent for several samples. The data on mobility can be fitted down to 86°K by an expression of the form $\mu = 2.54 \times 10^5 T^{-\frac{1}{2}}$, which would suggest interaction of the electrons mainly with acoustic vibrations of the lattice. However, there exists the possibility of scattering by impurities or other imperfections which in combination with optical scattering might also lead to the observed results. Strains play an important role in this material and are shown to be associated both with shallow 0.1-eV traps and with deeper traps. The average energy for production of one electron-hole pair by beta rays is 7.5 ± 0.5 eV. The range of holes in the samples tested, from 86°K to 150°K, was at least less than $\frac{1}{10}$ that of electrons.

I. INTRODUCTION

SEVERAL important advances have recently been made in the theory of the behavior of slow electrons in polar substances.¹⁻⁵ It has been emphasized³ that the results of this theory are applicable only for temperatures considerably below the Debye temperature corresponding to longitudinal optical vibrations, which is from 200 to 370°K for most ionic crystals. Except for the recent work of Redfield on Hall mobility in the alkali halides,⁶ there have been few experiments carried out at low temperature to compare with theory. Haynes and Shockley⁷ have made measurements near room temperature of Hall and drift mobility in single crystal AgCl. They have shown that in properly prepared samples the mean free time before trapping of photoelectrons is several microseconds, giving rise to long drift paths in an applied electric field. Similar long electron ranges at low temperature make it possible to detect single ionizing particles with the AgCl crystal counter,⁸ whereas in NaCl the ranges are quite short.⁹ AgCl, with a Debye temperature of about 280°K, is a convenient material upon which to make measurements of mobility at low temperature. In addition, interest centers on the silver halides in connection with the photographic process and properties of the solid state.¹⁰

The early crystal counter work with AgCl did not

yield characteristic or even very reproducible values of electron mobility.¹¹ However, by measuring rise times, Yamakawa¹² arrived at comparable values of mobility in two samples of AgBr at 77°K. Using improved crystal counter techniques, we will show that it is possible to get consistent results on AgCl provided that trapping and the effect of strains in the crystal can be minimized. Preliminary results have been given in a letter comparing low-temperature Hall and drift mobility in AgCl.¹³

II. THEORY OF THE METHOD

Principles of operation of the conductivity crystal counter have been treated by several authors.¹⁴ We are interested here primarily in the shape of the conductivity pulses. The annealed crystal of AgCl with shorted electrodes is slowly cooled to at least -75°C to minimize ionic conductivity. As shown in Fig. 1, a collecting voltage V_c can then be applied to the parallel electrodes (a) and (b) through an isolating resistance R . The capacitance, C , represents the combined capacity of crystal, leads, and preamplifier input. Ionization in the form of electron-hole pairs is produced near (a) by single beta particles which impinge upon the counter from below and stop in the crystal. With the top electrode (b) positive, the secondary or conduction electrons are drawn into the crystal, inducing a small charge, q , on the capacitance, C . Under conditions of large input

† Partially supported by grants from the National Science Foundation and the Research Corporation.

¹ H. Fröhlich and N. F. Mott, Proc. Roy. Soc. (London) **A62**, 446 (1949).

² Fröhlich, Pelzer, and Zienau, Phil. Mag. **41**, 221 (1950).

³ F. Low and D. Pines, Phys. Rev. **91**, 193 (1953); also Phys. Rev. **90**, 297 (1953).

⁴ E. P. Gross, Technical Report 55, Laboratory for Insulation Research, Massachusetts Institute of Technology, 1952 (unpublished).

⁵ F. Seitz, Revs. Modern Phys. **26**, 7 (1954); see pp. 23-24.

⁶ A. G. Redfield, Phys. Rev. **94**, 537 (1954).

⁷ J. R. Haynes and W. Shockley, Phys. Rev. **82**, 935 (1951).

⁸ P. J. van Heerden, thesis Utrecht, 1945 [see Physica **16**, 505 (1950)].

⁹ H. Witt, Z. Physik **128**, 442 (1950).

¹⁰ F. Seitz, Revs. Modern Phys. **23**, 328 (1951).

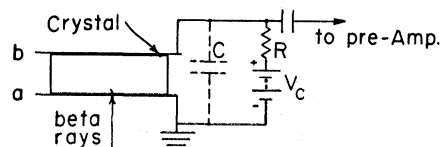


FIG. 1. Basic crystal counter circuit.

¹¹ R. Hofstadter, Nucleonics **4**, 4, 2 (1949); **4**, 5, 29 (1949).

¹² K. A. Yamakawa, Phys. Rev. **82**, 522 (1951).

¹³ F. C. Brown, Phys. Rev. **92**, 502 (1953); see also Phys. Rev. **92**, 858(A) (1953).

¹⁴ A. G. Chynoweth, Am. J. Phys. **20**, 218 (1952). See also references 8 and 11.

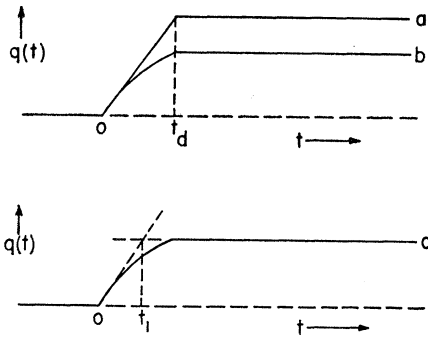


FIG. 2. Theoretical pulse shapes. *a*—for trapping unimportant, *b*—uniform distribution of traps, and *c*—showing the time t_1 .

time constant, a corresponding voltage pulse $v=q/C$ is produced at the amplifier input. Assuming direct recombination unimportant,¹⁵ the fate of the secondary electrons is that they either become trapped within the volume of the crystal or that they are collected at the top electrode. Holes, on the other hand, remain localized near the region of ionization, or if they have mobility, drift a short distance to the lower electrode. Reversal of the battery polarity making the top electrode negative allows one to look for pulses of opposite sign due to the drift of holes.

The time dependence of the charge pulse, q , is derived making the following simplifying assumptions: (1) that n_0 electron-hole pairs are all released instantaneously at time $t=0$ at the bottom electrode, $x=0$; (2) that a uniform electric field, $E=V_c/d$ exists in the insulating crystal where d is the sample thickness; and (3) that there is a uniform density of electron traps in the volume of the crystal giving rise to an average time, T , before trapping. The average distance the conduction electrons drift before trapping is called the *Schubweg* (displacement distance), w , and is given by $w=\mu ET$ where μ is the electron mobility (assumed a constant). Processes such as thermal release from traps are not considered at this time. The number of electrons still free to move a time t after release of the ionization is

$$n = n_0 e^{-t/T}. \quad (1)$$

The amount of charge induced on the electrodes by the drift of n electrons a small distance dx is

$$dq = ne \, dx/d. \quad (2)$$

With the aid of the substitution $dx = \mu E dt$, the charge $q(t)$ is found by integrating dq from 0 to time t .

$$q(t) = \frac{n_0 e \mu E}{d} \int_0^t e^{-t'/T} dt'. \quad (3)$$

¹⁵ The AgCl counter has been shown to have pulse-height response proportional to energy loss for particles ionizing near minimum, up to slow heavily-ionizing particles such as mesons stopping in the crystal. See H. G. Voorhies and J. C. Street, *Phys. Rev.* **76**, 1100 (1949). This proportionality fails for 5-Mev alpha particles, but this is believed due to the small penetration of the particles and a surface defect.

Expressing this in terms of w , we have

$$q(t) = (n_0 e w/d)(1 - e^{-\mu E t/w}), \quad (4)$$

which is applicable for times $t \leq d/\mu E$. For times greater than this the total charge induced is:

$$Q = (n_0 e w/d)(1 - e^{-d/w}). \quad (5)$$

A formula of this form was first derived by Hecht,¹⁶ who also showed that it could be verified experimentally for electrons released by light. The effect of finite penetration of the beta rays can be taken into account by subtracting the average penetration from d in the exponent of the exponential. For the case that trapping is less important and w is very large so that $\mu E t \ll w$, Eq. (4) becomes

$$q(t) \simeq (n_0 e \mu E/d)t. \quad (6)$$

Here the charge pulse has an approximately linear rise until a time $t_d = d/\mu E$ corresponding to collection of the n_0 electrons. In theory, a measurement of the time, t_d , allows one to compute the drift mobility from the relation

$$\mu = d/E t_d. \quad (7)$$

We shall show that this is also feasible in practice. It should be noted that even in the case that electron trapping is important, a break or discontinuity occurs at time t_d provided only that a fraction of the electrons reach the top electrode. A sketch of theoretical pulse shape is shown in Fig. 2. If one has an amplifier rise-time sufficiently short to make the detail of the pulse observable, all that is required to measure μ is a w long enough to approach saturation (collection of the majority of the secondary electrons). Mobility experimentally determined with the aid of Eq. (7) should be constant and independent of electric field strength; this condition, together with the observed pulse shape, serves as a check on the theory and its assumptions.

In the case that electrons are permanently trapped, a study of the shape of the pulses from an unsaturated crystal allows one to estimate the *Schubweg*, w . One convenient measure of the pulse shape is in terms of the quantity t_1 which is found from the intersection of the initial slope and the extrapolated final pulse height [see Fig. 2 (c)]. Defining t_1 in this way, $Q = q'(0)t_1$, where $q'(0)$ is the slope of the pulse at time $t=0$. The approximation used in Eq. (6) holds for the very beginning of the rise, so $q'(0) = n_0 e \mu E/d$. Multiplying this by t_1 and using Eq. (5) we arrive at an expression independent of n_0 :

$$w(1 - e^{-d/w}) = \mu E t_1. \quad (8)$$

Values of w obtained from an approximate solution of this equation can be compared with those obtained by the usual technique of fitting pulse height *versus* electric field data to the theoretical curve given by Hecht,¹⁶

¹⁶ K. Hecht, *Z. Physik* **77**, 235 (1932).

The above analysis could not be applied if temporary trapping of secondary electrons were important. This would be indicated by unusually long pulse rises and low values of mobility as computed from Eq. (7). In this case, the observed collection time, t_0 , would be more difficult to measure but it could be taken as the time for which the pulse approaches within a small fraction of its maximum value. It is given approximately by

$$t_0 = (d/\mu E) + t_i, \quad (9)$$

where t_i is the average time electrons spend in shallow traps and μ is the mobility of an electron in the conduction band. We shall return to this point in Sec. IV B.

III. EXPERIMENTAL APPARATUS

A. Crystal Growth and Sample Preparation

As is evident from the preceding discussion, it was desired to work with crystals in which the electron range, w , at low temperature is at least as great as the crystal thickness, about 0.5 cm. This implies well-annealed, relatively perfect crystals and high field strengths. It was discovered in earlier experience with silver chloride^{17,18} that crystals meeting these qualifications were not commercially available. Consequently the samples for the present experiments were grown from the melt¹⁹⁻²¹ by taking advantage of the purification which takes place during crystallization and allowing the history of each sample tested to be known. Raw material was cut from large pieces of unrolled crystal supplied by the Harshaw Chemical Company. This was melted in air at temperatures not exceeding 25° above the melting point in cylindrical, flat-bottom, platinum-lined crucibles²² which measured 1.5 cm in diameter and 7.6 cm in length. A crystal was then formed by slowly lowering the crucible and its contents through a steep temperature gradient in a furnace carefully stabilized by means of proportional control regulators. The rate of lowering was 1 mm per hour, and following solidification, the furnace and its contents were slowly reduced to room temperature over a period of 24 hours. The crystals could then be melted out by inserting the inverted crucible into a furnace set a few degrees above the melting point.

Disks of the order of 0.5 cm thick were cut from the crystal boules with a circular milling saw. Their surfaces were ground smooth and etched in sodium thiosulphate to remove metal or abrasive particles. Before the counting tests the samples were heat-treated after the manner of Haynes.²³ For this purpose we have found it

satisfactory to rest the crystals on special reagent-grade MgO within a steel-lined copper box in the heat-treating furnace. After annealing, the crystals were handled very carefully to avoid the introduction of strains due to thermal gradients or cold working, and all manipulation was carried out in the dark or under red safelight. Thin high-conductivity electrodes were applied to the parallel surfaces of the crystals using silver-conducting paint.²⁴

B. Optical Properties of the Crystals

Inspection under red polarized light indicated that the crystals were single and unstrained. Absorption of light in the tail of the fundamental band was compared for several samples after they had been used in the mobility experiments. The curve drawn through the open circles of Fig. 3 is typical of the absorption data taken with a Beckman Model DU spectrophotometer. The contribution to optical density due to successive reflections at the surfaces, but neglecting absorption of the reflected light, is shown as a dashed line in this figure. This reflection loss was computed by using the indices of refraction for AgCl given by Tilton *et al.*²⁵ Little variation in optical absorption was found between the four different crystals tested, including one sample cut directly from a large Harshaw crystal in which the Schubweg for electrons was of the order of one-tenth that in the recrystallized samples. It was not surprising to find the electronic properties much more sensitive to differences in composition or history of the crystals.

It should be emphasized that the low values of optical density for the long wavelengths shown in Fig. 3 (curve with open circles) were obtained only if the surfaces were very well polished and the crystal previously unexposed to blue light. Previous exposure or

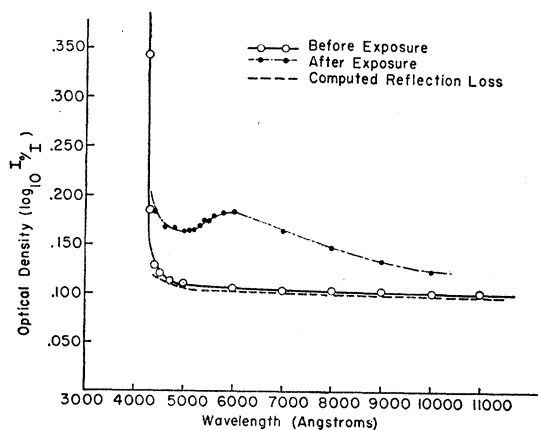


Fig. 3. Absorption of light in AgCl crystal II₂, thickness 0.65 cm, shown by the lower curve drawn through the open circles. The upper curve (solid dots) shows the darkening produced by long exposure to light of 4000 Å.

¹⁷ F. C. Brown and J. C. Street, *Phys. Rev.* **84**, 1183 (1951).

¹⁸ W. L. Whittemore and J. C. Street, *Phys. Rev.* **76**, 1786 (1949).

¹⁹ P. W. Bridgman, *Proc. Am. Acad. Arts Sci.* **60**, 305 (1925).

²⁰ D. C. Stockbarger, *Rev. Sci. Instr.* **7**, 133 (1936).

²¹ R. H. McFee, *J. Chem. Phys.* **15**, 856 (1947).

²² Obtained from Metals and Controls Corporation, General Plate Division, Attleboro, Massachusetts.

²³ J. R. Haynes, *Rev. Sci. Instr.* **19**, 51 (1948).

²⁴ Microcircuits Corporation, New Buffalo, New York.

²⁵ Tilton, Plyler, and Stephens, *J. Opt. Soc. Am.* **40**, 540 (1950).

surface properties may explain the slightly different results obtained by Gilileo²⁶ in the long-wavelength region. The effect of exposure of our crystals at room temperature to light of 4000 Å from the spectrophotometer for 10 minutes is shown by the solid dots. The crystals were not observed to darken very rapidly and there was evidence for saturation of the darkening in the molecular range as observed by Löhle.²⁷

C. Crystal Counter Apparatus

For the mobility measurements the crystals were mounted in the cryostat outlined in Fig. 4. The crystal rests on a brass shelf which is supported by a stainless steel tube forming the inner section of a Dewar. To slow down the rate of cooling, and to provide for intermediate temperature, the liquid nitrogen was sometimes poured into an inner cylindrical reservoir (not shown) which was supported and insulated by a piece of styrofoam. To aid in maintaining thermal equilibrium between the crystal and shelf, the closed space immediately surrounding the crystal was filled with dry nitrogen gas under pressure. A small-diameter thin-wall stainless steel tube leads from this space to the outside for evacuation and filling. Contact between crystal and shelf when in vacuum was not found adequate in the earlier runs, even though a drop of Silicone oil was

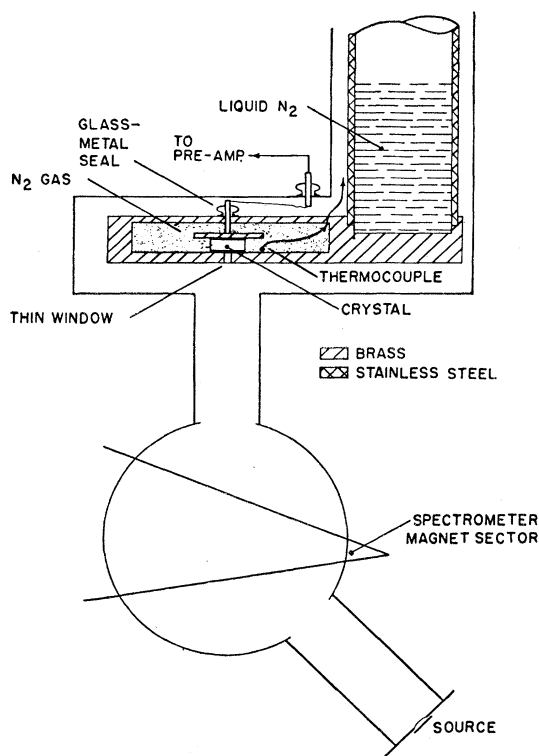


Fig. 4. Outline of the cryostat and beta-ray spectrometer.

²⁶ M. A. Gilileo, Phys. Rev. **91**, 534 (1953).

²⁷ F. Löhle, Göttingen Nachr. **2**, 271 (1933).

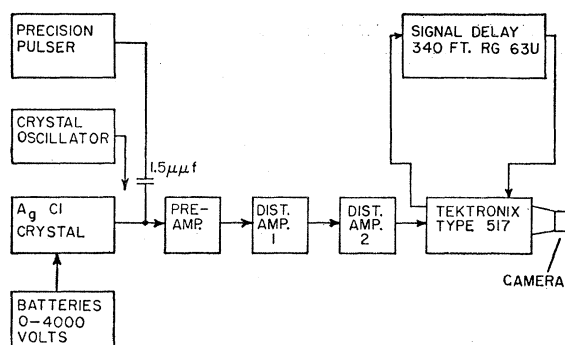


Fig. 5. Block diagram of electronic circuits.

placed at the interface. One of the difficulties here was that, because of the introduction of strains, the top electrode could bear only very lightly on the crystal. So that the crystal temperature could be related to the shelf temperature, a small copper-constantan thermocouple was imbedded in a hole in a test crystal, and measurements were taken. Vacuum pumps maintained a pressure of less than 10^{-4} mm of mercury in the space surrounding the shelf and liquid nitrogen reservoir.

A simple beta-ray spectrometer utilizing wedge-shaped magnet pole pieces and incorporating double focusing²⁸ was used as the source of ionizing particles. Such a design had the advantage of simplicity, ease of construction, good shielding against gamma radiation, and the location of the source and detector (the crystal) outside of the magnetic field. It had the disadvantages of relatively small solid angle and limited resolution. A $Ru^{106}-Rh^{106}$ source allowed the selection of roughly monoenergetic electrons from a few tenths of a Mev to over 3.0 Mev. Calibration at one energy was achieved by means of the internal conversion line of Cs^{137} .²⁹ For this purpose, and also to observe the counting efficiency of the crystals, a thin-window Geiger counter could be installed in place of the crystal above the hole in the brass shelf.

A block diagram of the crystal-counter circuits is shown in Fig. 5. A single beta particle releases only about 10^{-14} coulombs of charge in the crystal, and it is required that the amplifier faithfully pass the detail of a pulse which has a rise-time of as little as $0.10 \mu\text{sec}$. With the exception of the preamplifier, distributed amplification was employed throughout. A low-noise preamplifier circuit was developed which matched the high impedance of the crystal to the 200-ohm impedance of the distributed amplifiers. Its first stage employed a cascade-connected 6BQ7, which proved to be more stable, and to have lower input capacity than the more conventional arrangement. An over-all rise-time was attained of $0.018 \mu\text{sec}$. The clipping time of the system was greater than 10 microseconds so that slower rising pulses could be studied, although usually in this case

²⁸ W. G. Cross, Rev. Sci. Instr. **22**, 717 (1951).

²⁹ L. M. Langer and R. D. Moffat, Phys. Rev. **78**, 74 (1950).

the bandwidth was reduced with an improvement in signal to noise ratio. Amplitude calibration was achieved by applying a known amount of charge at the pre-amplifier input through a 1.5- μmf series capacitor. The timing calibration of the Tektronix-type 517 oscilloscope was checked with a crystal oscillator. Oscilloscope traces were photographed on 35 mm film and the pulses analyzed after projection.

IV. RESULTS AND DISCUSSION

A. Electron Drift Mobility

Figure 6 shows a group of pulses due to 2.0-Mev beta particles incident upon crystal number V_1 when the crystal temperature was 105°K. The pulses illustrate the general conditions under which the mobility data were taken for all the samples reported in Table I. Amplifier rise-time was of the order of 0.018 μsec and the voltage gain was adjusted so that 1 cm of pulsed height corresponded to 2×10^{-14} coulombs of charge at the crystal. For the first group of pulses on the left, the field was 9755 volts/cm and the sweep-time 0.110 μsec per cm. The essential features of the simple theory outlined in Sec. II are verified. An approximately linear rise indicates that the crystal was operating under nearly saturation conditions, as was verified also by little change in pulse height at different field strengths. Pulses with a lower field of 2250 volts/cm are shown below the test pulse on the right. Amplifier gain was the same as before but sweep-time was changed to 0.525 $\mu\text{sec}/\text{cm}$.

From ionization energy-loss considerations a 2.0-Mev electron should penetrate AgCl a distance of about 2.0 mm; a 0.5-Mev electron should penetrate only about 0.5 mm. However, the average penetration is less than this due to scattering of the beta particle, and the important quantity for purposes of computing pulse height is the center of gravity of the ionization as measured from the bottom electrode. For 3.0-Mev electrons the pulses became slightly rounded near the top of the rise, but less effect of this sort was noticed at lower energies where the rise-times were measured.

The results of a study of a large number of pulses for various energies selected by the spectrometer are in agreement with those of Van Heerden,⁸ who obtained approximate linearity of pulse height with energy for beta rays. For less penetrating radiation such as alpha

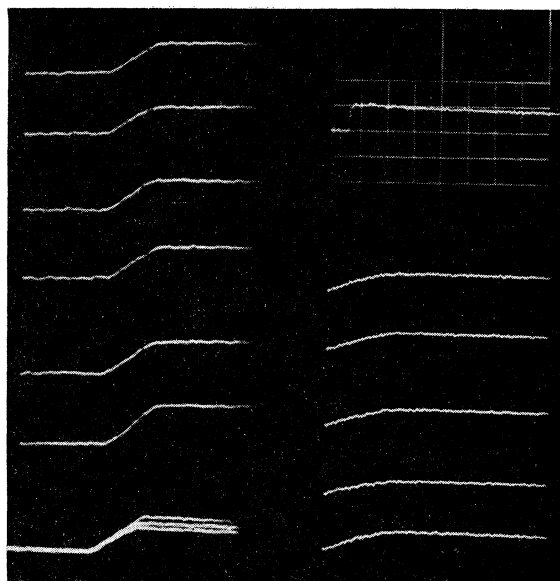


Fig. 6. Examples of crystal counter pulses produced by 2.0-Mev beta particles. At the left the field $E=9755$ volts/cm, the sweep time, 0.110 $\mu\text{sec}/\text{cm}$. A graticule ruled with 1-cm squares is shown on the right and superimposed is a test pulse of 2×10^{-14} coulomb. Below on the right $E=2250$ volts/cm and sweep-time, 0.525 $\mu\text{sec}/\text{cm}$.

particles, there seemed to exist a defective surface layer which sometimes could be observed for low-energy beta particles. When this condition existed, strains were visible under polarized light near the lower electrode. The condition could be improved by reannealing and then slow cooling to low temperature with less pressure of the top electrode on the crystal. In a properly cooled crystal (taking four hours or longer to reach -75°C), it was possible to demonstrate that these defective layers were at least less than 0.2 mm.

Electron mobility computed with the aid of Eq. (8) is tabulated for several crystals in Table I. The following procedure was used in arriving at the values of $\bar{\mu}$ given in the last column. Measurements were made of the time, t_d , for 50 to 100 pulses recorded at each condition of electric field and temperature. At high fields it was found that there was little scattering of the values of t_d from pulse to pulse. For example, in the most studied crystal, number V_1 , the value of the average of t_d for a group of data was 0.149 μsec at 86°K and 9755 volts/cm, whereas the root-mean-square deviation in t_d was 0.009 μsec or about 6 percent. At high fields the deviation was about what would be expected due to random noise on the pulse; at lower voltages, more variation occurred. Using the average of t_d for a group of pulses, mobility was computed from Eq. (8) and the results plotted as a function of electric field. Examples of the dependence of μ on E for crystal V_1 are shown in Fig. 7. It is seen that the values of μ become constant at high fields. Finally, the mobility $\bar{\mu}$ listed in Table I are the means of these nearly constant high-field values.

TABLE I. Electron mobility and range in annealed AgCl at low temperature. Sample H was cut directly from stock material without recrystallization.

Sample	Thickness d (cm)	w/E (cm ² /volt)	Temperature (°K)	Mobility, $\bar{\mu}$ (cm ² /volt sec)
V_1	0.408	8.2×10^{-5}	86 ± 3	274 ± 13
IV_1	0.450	1.1×10^{-4}	86	267
IV_2	0.462	1.4×10^{-4}	86	300
II_1	0.650	2×10^{-4}	86	268
H	0.570	1×10^{-5}	130	

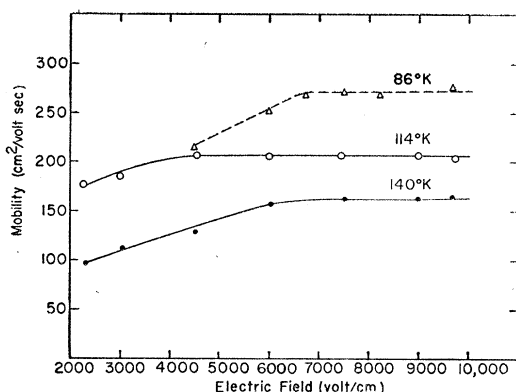


FIG. 7. Dependence of μ on E for crystal V_1 . The data for 86°K shows the effect of strains introduced by too rapid cooling from 114°K.

Crystal number V_1 was the most carefully studied. The error in $\bar{\mu}$ was estimated from the deviations in t_d and the other quantities to be less than ± 5 percent. The temperature in this case was known to within $\pm 3^\circ\text{K}$. Actually three separate runs were made on this crystal (each after annealing) with good reproducibility. Mobilities for the other crystals were more approximate and the temperatures less accurately determined. However, again some of the crystals were tested several times with consistent results.

The quantity w/E listed in Table I was estimated by fitting saturation curves of the type given by Eq. (6) to pulse height *versus* field data. In some cases where deep traps were predominant, a crude check could be had from the pulse shapes using Eq. (9). For the good crystals, w at a given field was not a constant for different runs, but depended upon the conditions of strain of the crystal.

Although the pulses at high field have a linear rise in agreement with theory, two objections may be brought up concerning the use of Eq. (5) and the simple theory from which it was derived in computing mobility: (1) It might be stated that the existence of a volume distribution of charge such as produces polarization of a crystal counter may cause the internal field to be different than given by $E = V_c/d$; and (2) the distance the secondary electrons move may not be d , the thickness of the crystal, due to boundaries or trapping layers in the volume of the crystal. That polarization effects were not important in our case can be deduced from the following arguments. Silver chloride at room temperature is an ionic conductor which, in the absence of an externally applied electric field, should be charge neutral throughout its volume. The crystals were cooled with shorted electrodes to low temperature, where the ionic conductivity is negligible. The collecting voltage was then applied and *single particles* detected at low counting rates, usually of the order of 100 counts/minute. Any variation of the pulse height could be observed as the experiment progressed. Moreover, using a gamma ray source, a test could be made for the

existence of internal fields due to bound charge by removing V_c and looking for pulses of opposite polarity. Polarization effects did not begin to become important until about 10^4 to 10^5 counts, which was more than registered during the runs. This is in agreement with the results reported by others for AgCl.³⁰ We have here one of the chief advantages of the method as compared with bombardment-induced conductivity techniques in which polarization effects are certainly more important.³¹

That trapping layers were not important is shown by the consistent values of mobility obtained from crystal to crystal, and for a crystal operated first in one position and then inverted. In addition, early tests were made in which the crystal could be moved around on the brass shelf without finding very marked variations over the crystal area. An important consideration bearing on these matters is the average energy for production of one electron-hole pair, a quantity which we will call ϵ . For the mobility runs, ϵ was of the order or less than 10 electron volts per ion pair. It was carefully measured for crystal V_1 by setting the spectrometer to the internal conversion line of Cs^{137} . A pulse-height distribution for a total of 550 pulses is shown in Fig. 8. This is in agreement with previous results⁸ where it was argued that the distribution of smaller pulses was primarily due to the scattering of the beta particles. The peak of the distribution corresponds to 10^{-14} coulombs of induced charge. After making small corrections for window thickness, lack of saturation of the crystal and particle penetration one gets a value of $\epsilon = 7.5 \pm 0.5$ electron volts/ion pair.

The dependence of drift mobility on temperature is shown in Fig. 9, which summarizes data for three crystals and shows also the higher-temperature points given by Haynes and Shockley.⁷ The mobility measurements of Fig. 9 are accurate to within about 10 percent for crystals II, and IV, and within 5 percent for

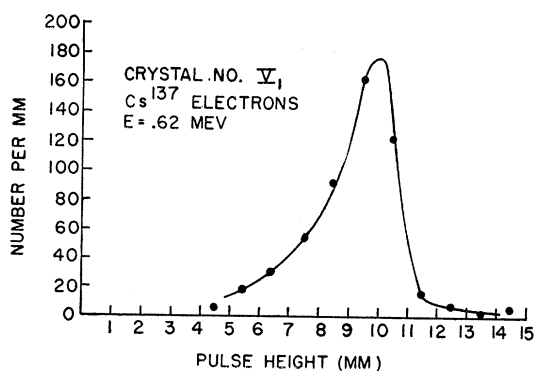


FIG. 8. Pulse height distribution for monoenergetic, 0.62-Mev beta rays. A maximum near 10 mm corresponds to 10^{-14} coulombs and yields a value of $\epsilon = 7.5 \pm 0.5$ ev/ion pair.

³⁰ L. F. Wouters and R. S. Christian, U. S. Atomic Energy Commission document MDDC-1324, 1947 (unpublished).

³¹ K. G. McKay, Phys. Rev. **74**, 1606 (1948); and **77**, 816 (1950).

crystal V, with an uncertainty in temperature of, at most, $\pm 5^\circ\text{K}$. Data for only three crystals are given, but it is believed that the results are representative of μ versus T for AgCl. The values at low temperature are in agreement with the early crystal counter work on AgCl; for instance, Hofstadter cites values from 114 to 400 $\text{cm}^2/\text{volt sec}$ at 77°K . Moreover, in view of the constancy of μ vs E at high fields, it is reasonable to assume that the drift mobility we have measured is close to the so-called microscopic mobility in AgCl.⁷ In this connection note that Hall mobility¹³ measured from the Hall angle (not corrected by $8/3\pi$) in one of our samples at 86°K lies just above the drift mobility.

Let us first compare the data with the theory of Low and Pines,³ who assume that electrons are scattered primarily by optical modes of vibration, as was also assumed in the earlier work of Fröhlich and Mott.⁴ They give an expression which is applicable only for $T \ll \Theta$, as follows:

$$\mu = \frac{1}{2\alpha\omega} \frac{e}{m} \left(\frac{m}{m^*}\right)^2 f(\alpha)(e^{\Theta/T} - 1), \quad (10)$$

where we have subtracted 1 from the exponential to give the proper convergence at higher temperatures. The constant α measures the strength of the electron-lattice interaction and is proportional to $m^{\frac{1}{2}}$; $f(\alpha)$ is a slowly varying function about 1.14 in our case; m is the effective mass of the electron in the conduction band and, since this is unknown, plays the role of an adjustable parameter in the theory; and finally, the effective mass of electron plus associated lattice polarization is given by $m^* = m(1 + \alpha/6)$. The Debye temperature $\Theta = \hbar\omega/k$ corresponds to the frequency, ω , of the longitudinal optical modes of vibration of the lattice. Using $\omega = \omega_L(\epsilon/n^2)^{\frac{1}{2}}$, we can deduce Θ from the low- and high-frequency dielectric constants, $\epsilon = 12.3$, $n^2 = 4.04$,³² and the reststrahl wavelength $\lambda_L = 90$ microns.³³ We arrive at $\Theta = 280^\circ\text{K}$ for AgCl. Now if the experimental points for μ below about 140°K are plotted on a semilog basis against $1/T$, it is quite clear that they do *not* lie along an $e^{\Theta/T} - 1$ curve (rough agreement can be had by assuming an unreasonably low Debye temperature $\Theta \approx 100^\circ\text{K}$).

On the other hand, the data are seen to agree quite well over a wide temperature range with a $T^{-\frac{1}{2}}$ law as would be predicted by scattering with the acoustical modes of vibration.³⁴ The solid line of Fig. 9 is given by

$$\mu = 2.54 \times 10^5 T^{-\frac{1}{2}}, \quad (11)$$

which was shown by Haynes and Shockley to fit their results. If, in order to match the $T^{-\frac{1}{2}}$ data, we assume that the interaction over the entire temperature range

³² K. Hojendahl, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 16, No. 2 (1938).

³³ R. B. Barnes, Z. Physik 75, 723 (1932), extrapolating for AgCl between the points given.

³⁴ A. H. Wilson, *The Theory of Metals* (Cambridge University Press, London, 1953), p. 265.

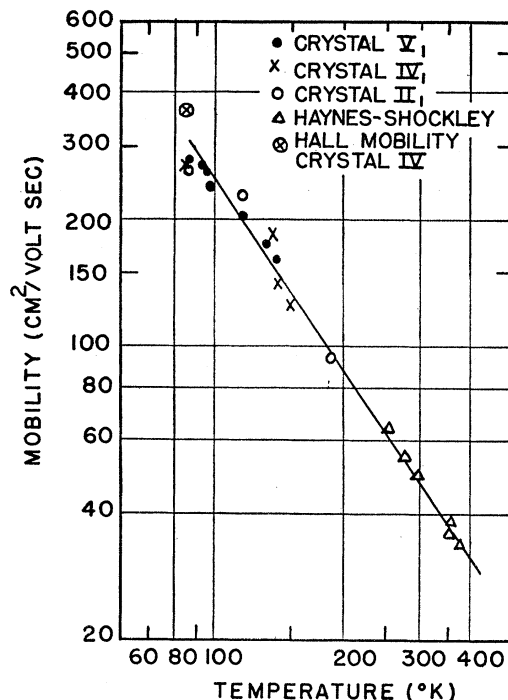


Fig. 9. Mobility as a function of temperature for electrons in AgCl.

is primarily acoustic, and, at the same time, that the theory based on optical modes is applicable below say 140°K , it is necessary to pick the coefficient of $e^{\Theta/T} - 1$ in Eq. 10 to be about 50 or larger. This corresponds to an effective mass $m \approx 0.2m_e$, where m_e is the free electron mass. Now, although the applicability of an acoustic equation, such as Wilson³⁴ derives, to an ionic crystal is not clear, we can roughly estimate mobility for $m \approx 0.2m_e$ and arrive at values which are quite a bit higher than the experimental points. It seems necessary to pick a larger effective mass for agreement with the acoustic theory.

There is an unresolved difficulty here. The theoretical situation, which is exceedingly complex, merits further examination. On the experimental side, it would be interesting to have more data at lower temperatures. It is conceivable that scattering by dislocations or other imperfections is important at 86°K , but in this case one would expect greater variation in mobility from sample to sample. A theory based on ionized impurity scattering along with optical scattering might explain the observed results, but a fairly high concentration of impurity atoms is required.^{35a}

³⁵ The work of C. Allemand and J. Rossel, *Helv. Phys. Acta.* 27, 212 (1954), has recently come to the attention of the author. It would appear that the composition of their crystals is quite different from ours.

^{35a} Note added in proof.—In a private communication D. Pines has pointed out an error in the expression for mobility given by reference 3. Equation 10 should contain the factor m/m^* raised to the third power rather than the second, that is, μ should be reduced by a factor m/m^* . This makes little change, however, in the above arguments.

B. Additional Observations

The conductivity pulses from our better crystals at low fields were rounded and had unusually long rise-times which gives evidence for trapping and delayed release of charge from very shallow traps. This situation resulted in a decrease in the observed μ at low E , as shown in Fig. 7. The effect was not noticeable at high fields, presumably because a smaller fraction of secondary electrons became trapped during the short times to cross the crystal thickness, d .

We may roughly estimate the depth below the conduction band for these shallow traps by first finding the average time, t_t , an electron spends in traps near the lower electrode. The observed collection time, t_0 , is given by Eq. (10) and to this corresponds an observed mobility $\mu_0 = d/Et_0$. Rearranging Eq. (10), we get

$$\frac{1}{\mu_0} = \frac{1}{\mu} + \frac{t_t E}{d}. \quad (12)$$

Thus t_t may be estimated from the slope of the $1/\mu_0$ vs E data at low E . For crystal V at 140°K we find $t_t \approx 0.5$ μ sec. Supposing that conditions are such that a negligible fraction of the electrons are trapped more than once, it is reasonable to associate this t_t with the average time an electron spends in a trap. Saying that the probability per unit time for escape from a trap $B = B_0 e^{-E/kT} = 2 \times 10^6$ sec⁻¹, the thermal trap depth, E , is about 0.1 electron volts, depending slightly upon what value of B_0 is chosen.³⁶ Traps of about this depth have previously been found in AgCl from the results of "glow curve" experiments.³⁷

It is quite clear that strains play an important role in affecting the range of electrons in AgCl. The concentration of both the shallow, 0.1 eV, traps and of deeper traps could be increased by too rapid cooling or by mild cold-working (such as produced by moving the crystal as it rested under the top electrode on the brass

shelf). There was evidence that these effects depended somewhat upon temperature; for example, a crystal was more resistant to the effects of strains while at the lowest temperature 86°K. Figure 7 shows how shallow traps became more important (μ dropping off at a higher E) in crystal V after rapidly cooling from 114°K to 86°K. Subsequent examination under red polarized light showed the presence of strains near the bottom electrode which could be removed as usual by heat treatment.

Simpson³⁸ has calculated the thermal trap depth of an interstitial silver ion in AgCl and arrived at about 0.1 electron volts. It is possible that cold-working increases the concentration of interstitial silver in our crystals. Such would be the case if Frenkel defects were generated in sufficient numbers by dislocations during cold work.³⁹

A search was made in all of the crystals for evidence that positive holes drift with ranges comparable to that of electrons. Hole pulses were not observed at temperatures from 86°K to 150°K. Small pulses of opposite polarity were observed under reversed field (top electrode negative), but only for the most penetrating beta particles. They could be explained in terms of sweeping of the secondary electrons toward the lower electrode. We are led to conclude that if holes have a mobility comparable with electrons, their range is less than $\frac{1}{10}$ that of electrons. We would not have been able to detect the migration of holes with very low drift velocity.

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³⁶ N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, London, 1940), p. 108.

³⁷ A. Moore, thesis, Cornell University, 1951 (unpublished).

³⁸ J. H. Simpson, Proc. Roy. Soc. (London) **A197**, 269 (1949).

³⁹ F. Seitz, *Advances in Physics* **1**, 43 (1952).

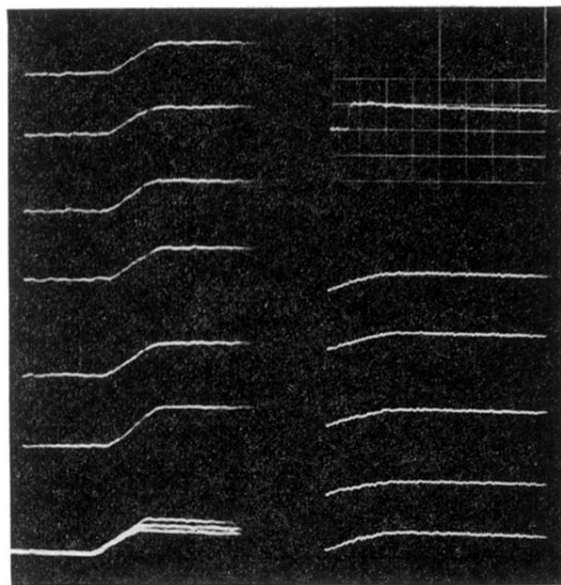


FIG. 6. Examples of crystal counter pulses produced by 2.0-Mev beta particles. At the left the field $E=9755$ volts/cm, the sweep time, $0.110 \mu\text{sec/cm}$. A graticule ruled with 1-cm squares is shown on the right and superimposed is a test pulse of 2×10^{-14} coulomb. Below on the right $E=2250$ volts/cm and sweep-time, $0.525 \mu\text{sec/cm}$.