Thallium halide radiation detectors

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During a series of experiments on crystal conduction counters performed at Stanford University on thallium halide crystals, we have observed motion of both hole and electron carriers in a TlBr crystal. At a temperature near −90 °C the hole motion produces larger pulses than electron motion. We have studied the behavior of TlBr, TlCl, and KRS-5 (40 mol % TlBr + 60 mol % TlI) crystals and examined them as possible crystal conduction detectors of α particles and γ rays. TlBr appears to be a promising candidate for applications to nuclear physics and high-energy γ-ray physics. Modules of TlBr in "crystal-ball" geometry may lead to new detection possibilities. At −20 °C space-charge accumulation in TlBr decreases to such an extent that operation at this temperature seems possible with moderate electrical gradients. In the long-neglected field of crystal conduction counters, we have potentially removed the space-charge limitation in TlBr and, allowing for both hole and electron motion, raised the possibility for spectroscopic performance of this material for γ-ray studies.

INTRODUCTION

The fact that several insulating solids have the property of detecting α, β, and γ radiation has been reported by many workers,[1–11] including van Heerden,[5,6] who was the first to report the counting phenomenon in solids. The search for compounds using high-Z elements that could possibly be used as detectors for nuclear radiation, especially γ rays, has caused attention to be focused on thallium halides for a long time. We note that KRS-5 (40 mol % TlBr + 60 mol % TlI) was reported by Hofstadter to possess this desirable property as early as 1947. The theory of such conduction counters has been dealt with by Hofstadter, and the whole subject has been thoroughly reviewed by him in a series of articles[1,2] published in 1949. The counting property depends on the prompt collection of electrons and/or holes that are produced in the solid substance by the passage through it of an elementary particle either from a radioactive source or from an accelerator. The "free" charges are swept towards the applied contacts of a crystal across which a suitably high electric field is maintained. If a charged particle moves a distance y parallel to the field E, the charge it induces on the electrodes is ey/d, where e is its charge and d is the separation of the electrodes (Fig. 1). The magnitude of the voltage pulse obtained, therefore, depends on the average distance $\bar{y}$ traveled by all the secondary carriers.

Unfortunately, the number of serious drawbacks from which such detectors have invariably suffered have slowed down the progress of research on the insulating crystal conduction counter. Firstly, a crystal that is used to count any type of particle develops an internal reverse electric field caused by the trapping of secondary charges inside the crystal whose movement is intended to give rise to a voltage pulse across the electrodes. The traps are impurity levels, line or point defects, or other structural irregularities that are more or less uniformly distributed in the body of the crystal. The space charge[1,10] that results from the trapping of the secondary charges (electrons or holes) sets up an internal electric field in a direction usually opposite to the applied field. The drift velocity of the ions is thereby reduced, which results in a still greater chance of capture by the traps. Sometimes the reverse field is called a "polarization" field.

The mean trapping distance δ, also known as the *Schubweg*, is related to the drift velocity v_d as

$$\delta = v_d T = \mu E T , \quad (1)$$

where μ is the mobility and T the average trapping time.[12] The trapping causes an exponential decrease in the number of moving charges given by

$$n = n_0 e^{-y/\delta} , \quad (2)$$

where n is the number that survive out of n_0 after having

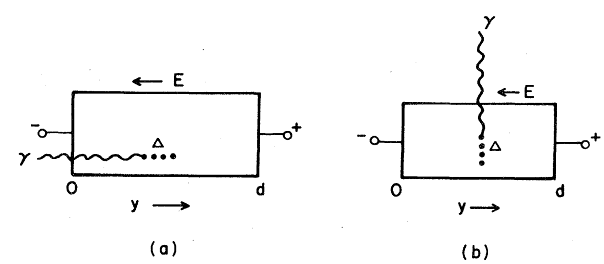

FIG. 1. (a) Applied electric field parallel to the beam direction, where Δ is the length of the ionized track and d is the crystal thickness. (b) Field perpendicular to the direction of the beam. d is the crystal thickness.

moved through a distance y in the crystal parallel to the applied field. This movement is, of course, a drift movement superposed on the random scattering of these particles. A number of remedies have been suggested by early workers for the removal of the space charge. A relatively high applied field will ensure a large value of δ, but there may be practical considerations such as breakdown so that a very high field cannot usually be used. The accumulated space charge can often be removed from the traps between counting periods by shining light of a suitable wavelength on the crystal. This method has been successfully used by the present authors.

Secondly, in many insulating crystals reported in the literature so far, except diamond at room temperature,[7] only one type of carrier, the electrons, has been shown to be responsible for the conduction current. The holes have usually been assumed to remain immobile. Although lack of hole mobility also gives rise to an additional space charge enhancing the effect caused by the trapping of electrons, its most serious effect is to jeopardize the energy resolution in case of γ rays. The energy resolution in case of monoenergetic α or β rays depends mainly on the inhomogeneities in the crystal, since every α or β particle will give rise to the same number of elementary charge carriers in the same part of the crystal, e.g., near the electrode through which the particles enter. However, γ rays can interact anywhere in the crystal. The Compton electron, pair electron, or photoelectron knocked out loses its energy by secondary ionization over a small distance (Δ) where the event takes place. The size or height of the pulse thus produced depends on the point of interaction. If, however, holes as well as electrons were mobile and losses due to trapping were small, the voltage pulses due to the charge carriers released by the photoelectrons would all have virtually the same magnitude irrespective of the point of interaction of the γ ray, and good energy resolution would be achieved.[13] The Compton electrons produced by monoenergetic γ rays have a rather wide energy and angular distribution and would give rise to a wide background much as is observed through the use of a NaI(Tl) scintillation detector.

The sensitivity and the efficiency of the conduction counter therefore depends on the ability of the incident particle to produce a large number of electrons and holes from the crystal atoms, and also on the ability of the positive and negative carriers so produced to move all the way from where they are created to the extremities of the detector, without being lost on their way, e.g., by trapping. The first factor in turn depends on the energy required to create an electron-hole pair, and this is directly related to the optical band gap E_g of the material used. The "energy per ion pair" or "energy per free electron" is related to the efficiency of production (Ψ) of the conduction electrons. If the incident particle has kinetic energy E, the number of ion pairs created is

$$n_0=(E/E_g)\Psi \ . \qquad (3)$$

As some energy is always wasted thermally in the form of excitons or phonons, the quantity Ψ is always less than unity. If $\Psi=1$, the number n_0 would have its maximum value controlled only by the optical band gap of the crystal material. The effective energy per ion pair is thus E_g/Ψ. van Heerden[6] finds a value of 7.6 eV for this quantity in crystals of silver chloride. Gases seem to have the largest value per ion pair of about 30 eV. For nitrogen this quantity is 36 eV. Again, in order that all the charges created by ionization are collected and give rise to a large output electrical signal, the quantity δ, the mean trapping distance, must be large.

We report here the work done and the results obtained from a study of crystals of thallium chloride, thallium bromide, and KRS-5, an eutectic mixture of 40 mol % TlBr and 60 mol % TlI. It is particularly emphasized that a large hole motion has been observed in TlBr crystals which we have not seen in other crystals.

II. EXPERIMENTAL

The apparatus used for studying the properties of the crystals is shown in Fig. 2. It consisted mainly of a stainless-steel vacuum chamber which was covered by an aluminum lid. The lid had a 7-in. deep and $1\frac{1}{2}$-in.-diam

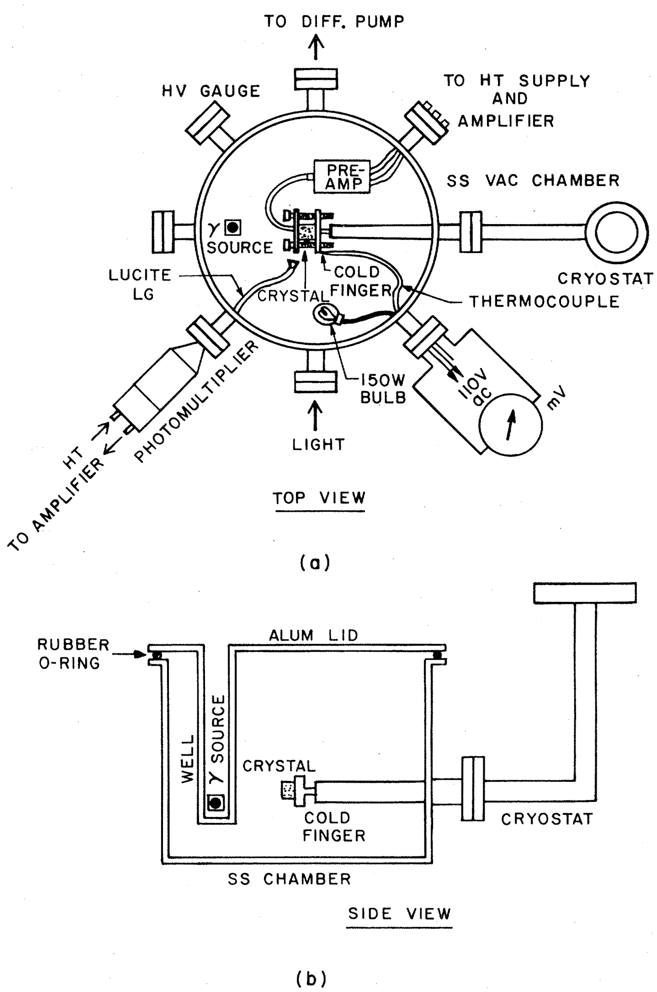

FIG. 2. (a) Shows the stainless-steel (SS) vacuum chamber, the liquid-nitrogen cryostat, the position of the crystal holder, and other pieces of apparatus. (HV denotes high vacuum, LG denotes light guide, and HT denotes high voltage.) (b) The side view shows more clearly the position of the aluminum well wherein the γ source was placed.

"well" attached to it in which a γ source could be inserted. The internal surfaces of the chamber were bright nickel plated so that light incident through one of the quartz window ports could be reflected towards the crystal. However, the crystal was usually kept in the dark, except when it was desired to remove polarization. The crystal itself was placed in thermal contact with a 2-in.-diam copper block (cold finger) cooled by liquid nitrogen and was clamped loosely to it by means of four nylon screws [Fig. 2(a)]. The crystals were obtained from the Harshaw Chemical Company and were 1 in. in diameter and 0.3—1.3 cm thick. Both plane faces of each crystal were coated by a 2000-Å-thick layer of gold either by sputtering or vacuum evaporation and served as electrical contacts. A thin circular copper ring placed over the front surface of the crystal served as the high-voltage contact. The back surface of the crystal was connected to ground through the copper block on which it was mounted. The preamplifier (Ortec model 118A) was placed inside the chamber close to the crystal in order to avoid pickup of stray electrical signals. The temperature of the crystal was monitored by a calibrated copper-Constantan thermocouple. A 150-W incandescent bulb was used to warm up the crystal when desired.

A block diagram of the measuring electronics is shown in Fig. 3. Pulses coming out of the preamplifier were amplified by a TC205A amplifier, and the output of the amplifier was fed into a linear gate stretcher (model 442, Ortec). The linear gate provided rectangular pulses of constant width whose height was proportional to the input pulse height (gain = 0.88). The output of the linear gate was fed into a multichannel analyzer to obtain the pulse-height distribution of the charge pulses produced by the crystal. The preamplifier used was a "charge-sensitive" type, so that each output pulse had an amplitude proportional to the charge induced on the ungrounded electrode of the crystal. The crystal capacitance ranged from 30 to 35 pF, whereas the feedback capacitance c_f in the charge-sensitive amplifier was about half a picofarad. A voltage gain of about 60—70 was thus provided by the charge-sensitive preamplifier.

The vacuum chamber had several ports, two of which had quartz windows. One of these ports was used for illuminating the crystal by light of any desired wavelength. The other window enabled us to couple the photocathode of an RCA 8850 photomultiplier tube to the crystal through a lucite light guide. This arrangement enabled us to look for scintillations.[14]

The chamber was maintained at a pressure of less than 10^{-5} Torr. A good vacuum was ensured before adding liquid nitrogen in the chamber so that water vapor or other contaminants did not condense on the crystal surfaces. The aluminum well had $\frac{1}{4}$-in.-thick walls and an internal diameter of 1.5 in. This thickness prevented the counting of low-energy β particles from γ sources. By rotating the lid through 90° it was possible to irradiate the crystal, if necessary, by γ rays in a direction perpendicular to the electric field. For irradiation by α particles from an ^{241}Am source we used a separate collimator, which was put in front of the crystal and had its shutter operated electromagnetically from outside.

Before taking any observations, the crystal was cleared of any space charge accumulated in the trapping centers. This was done by shining light through a suitable filter for each different type of crystal while the crystal was being bombarded by α or γ radiation with no applied field. Positive or negative (or both) pulses could be observed by the preamplifier if the space-charge field was strong enough, and they would usually disappear within a few seconds after shining light on the crystal.

RESULTS

Our results can be divided into several parts, namely (i) the energy resolution obtained with monoenergetic α particles, (ii) the effect of temperature on the energy resolution and the electron or hole pulse heights, (iii) the value of the energy per ion pair, (iv) the types of pulse-height spectra obtained by irradiation of the crystals at the optimum settings of the electric field and temperature by various γ-ray and α-particle sources, and (v) scintillating properties, if any. The space-charge effects are discussed later.

THALLIUM BROMIDE

In the following we discuss the results listed in the preceding section.

(i) The best energy resolution obtained with α particles from ^{241}Am was about 19% full width at half maximum (FWHM) at a temperature of -90 °C and an electric field of 2760 V/cm. The experimental resolution due to

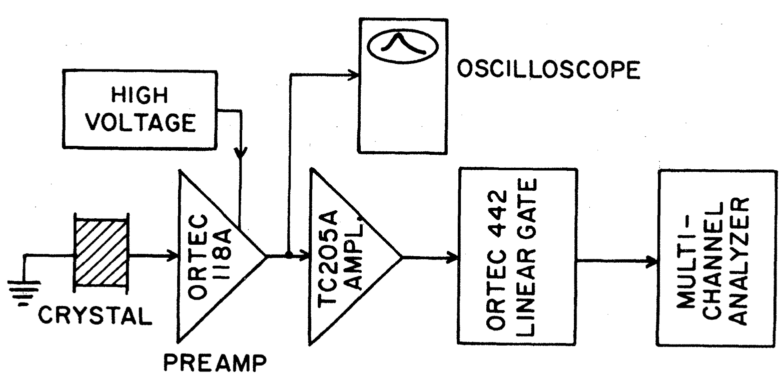

FIG. 3. Block diagram of the measuring electronics.

thermal noise from the preamplifier and due to statistical variations in the number of electron-hole pairs was less than 3%. The reason for the observed spread in pulse height is not known but may be due to a nonuniform surface layer and variations in the mean trapping distance δ in different parts of the crystal. The α-particle collimator had a channel whose diameter was 4 mm, and the α-particle beam fell over a circular area of about 6-mm diameter on the crystal surface. When the hole area was reduced to 2-mm square, the energy resolution did not improve significantly. A typical pulse-height spectrum is shown in Fig. 4. It may be noted that this spectrum was taken with a positive applied voltage of + 1300 V. Thus mainly holes were responsible for the production of the charge pulses whose amplitude distribution is shown here. This effect was noted only in TlBr and not in other thallium halide crystals. To our knowledge, this is the first time that a large movement of holes has been found to give rise to pulses in a thallium halide used as a conduction counter at a low temperature. The efficiency for counting α particles was close to 100%. The sample of TlBr used had an impurity content less than 20 ppm. (This estimate was given to us by the manufacturer, Harshaw Chemical Company.) It is to be noted that our results refer to the one and only crystal of TlBr we had in our possession.

(ii) The effect of varying the temperature on the most probable pulse height and the energy resolution for α particles from an ^{241}Am radio source is shown in Fig. 5, from which it appears that the optimum temperature for best energy resolution is about $-90\,^{\circ}$C, with an applied field of about 2000 V/cm. A slight improvement in energy resolution may be obtained by using a higher electric field. Incomplete charge collection at lower fields, however, is responsible for only a small percentage of the total spread in the pulse-height distribution. If the total number of charge carriers released by each α particle had the same value N, this will contribute an amount $\sim 1/\sqrt{N}$ to the

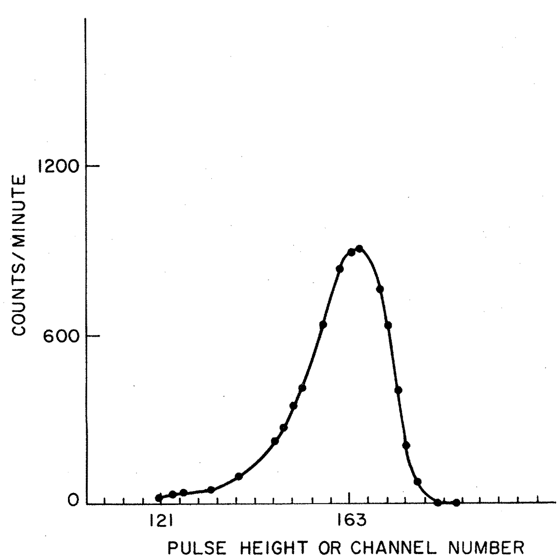

FIG. 4. ^{241}Am α-particle pulse-height spectrum. For details see text.

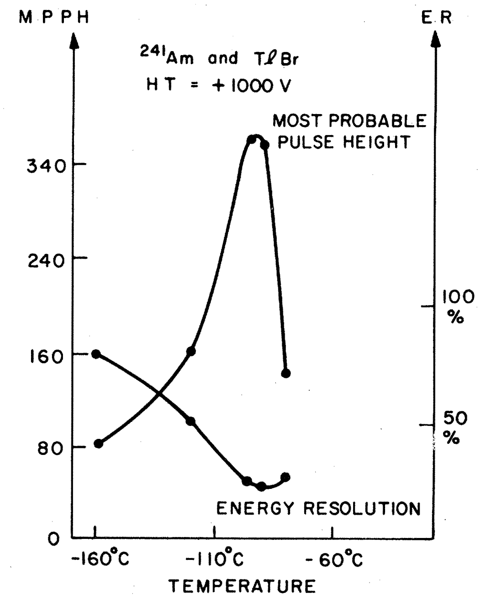

FIG. 5. Most probable pulse height (MPPH) and the energy resolution (ER, FWHM) for ^{241}Am α particles as a function of the crystal (TlBr) temperature.

energy resolution.[15] The large spread in the α-particle spectrum is not, therefore, well understood and perhaps a small part of it may be instrumental. Incidentally, with a crude CsI(Tl) scintillation detector the spread in the α-particle scintillation peak was nearly the same ($\simeq$13%) as that for the 0.57-MeV γ ray of ^{207}Bi, which indicated that the α-particle beam itself was reasonably monoenergetic. The variation in the pulse height with temperature may possibly be understood as follows. As the temperature is increased the holes spend less time in some of the trapping centers and tend to produce a larger pulse, whereas the hole mobility μ_h decreases with increased temperature, and this effect tends to reduce the pulse height: The combined effect obtained by multiplying the two factors may give rise to the observed maximum in the most probable pulse height.

(iii) The energy per ion pair calculated from the measured maximum pulse height produced by the 1.33- and 1.17-MeV γ rays of ^{60}Co at a temperature of $-105\,^{\circ}$C and an electric field of 3190 V/cm, and from the measured calibration of the charge-sensitive preamplifier, was found to be 19 ± 2 eV. The minimum in the band gap of TlBr probably occurs at 5000 Å or equivalently at 2.48 eV. The value of Ψ in this case as defined by Eq. (3) is therefore about 0.13. We obtain a much higher figure for the energy per ion pair if we use the maximum pulse height obtained from α particles. However, as van Heerden has suggested, the lower pulse height could be due to the creation of intense local fields that affect the movement of carriers away from the site of ionization. The results of Yamakawa[8,9] on α particles produced within the body of a mixed crystal of LiBr and AgBr by slow neutron capture showed that α pulses are larger than γ pulses of energy by about 2 MeV. Thus surface effects may be important in α-particle determinations of energy per ion pair.[16]

(iv) Pulse-height spectra obtained with ^{241}Am, ^{60}Co, and

^{207}Bi radioactive sources are shown in Figs. 4 and 6. The α-particle spectra are produced by the motion of holes created by ionization near the positive electrode through which the α particles entered. The shape of these spectra is as expected from van Heerden's theory. Our experimental resolution as measured with a pulser was only about 2%, and so the entire width of the spectrum may be due to surface effects as already suspected. ^{60}Co emits two γ rays of energies, 1.33 and 1.17 MeV. They interact mainly via Compton and photoelectric processes. The ^{60}Co spectrum shows a pronounced bump due to the overlapping of the photopeaks of the two γ rays and the Compton edge. By carefully measuring the negative and the positive pulse heights due to α particles with negative and positive high voltages applied in turn to the ungrounded contact, we determined that the negative electron pulses were approximately 35—45 % as large as the hole pulses. With only a small electronic contribution the photopeaks are smeared, and this gives rise to the wide bump in the middle. The sharp peak near zero pulse height is probably instrumental. The effect of varying the electric field on pulse height due to α particles is shown in Fig. 7. The rise time of α-particle pulses was about half a microsecond when the temperature was $-100\,^{\circ}$C and the voltage gradient nearly 2000 V/cm.

A ^{207}Bi spectrum taken with the crystal (TlBr) bombarded from the side [Fig. 1(b)] shows two small bumps at channels 100 and 192 of the multichannel analyzer. The ratio of the channel numbers is 1.92, which is very nearly the same as the ratio of the known energies of 1.06 and 0.57 MeV of the two most abundant γ emissions of ^{207}Bi. The peaks are more pronounced and resolved here than in the ^{60}Co case because of the higher photoelectric cross sections at the lower energies and the relatively large energy separation of the two γ rays. The Compton edge is also clearly visible.

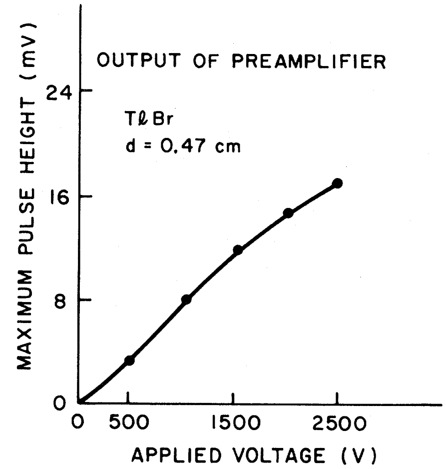

FIG. 7. Variation of maximum pulse height due to α particles from ^{241}Am as a function of the high voltage applied across the crystal.

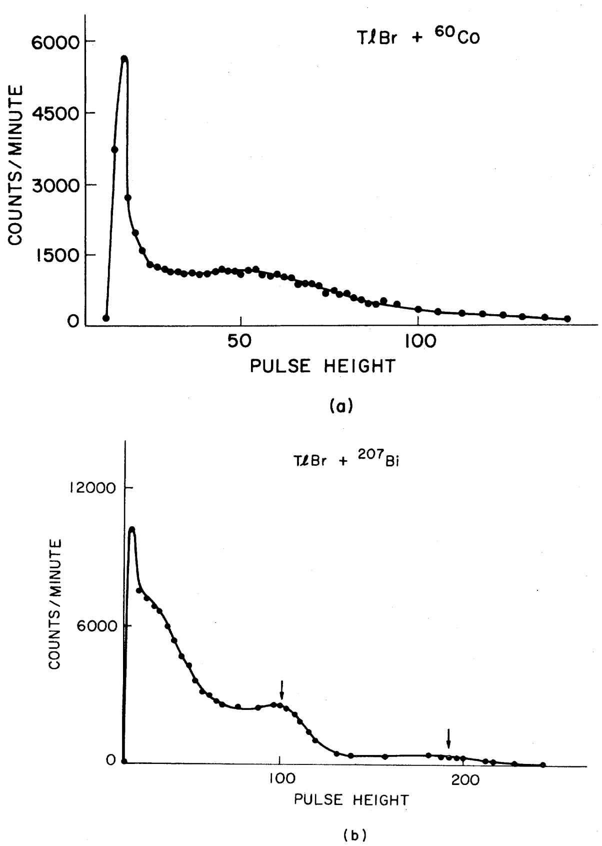

FIG. 6. (a) ^{60}Co γ-ray pulse-height spectrum produced by TlBr conduction counter. See text for details. (b) ^{207}Bi γ-ray pulse-height spectrum obtained with TlBr near the optimum temperature.

(v) No scintillation pulses could be detected either by α or γ bombardment of the TlBr crystal that was used. The yield of photons was smaller than 1% of the height of an equivalent NaI(Tl) pulse. The dielectric constant of TlBr sample used was measured and was independent of temperature, having a value of about 31.

HIGH-TEMPERATURE BEHAVIOR OF TlBr

In addition to being a useful counter at low temperatures, TlBr also shows promising results near a temperature of $-20\,^{\circ}$C. This temperature can be readily achieved without the use of liquid nitrogen. Pulses of 1.2 mV were observed with an applied field of 400 V/cm when the crystal was bombarded by α particles from ^{241}Am at a temperature of $-20\,^{\circ}$C. Pulses due to 1-MeV γ rays were nearly 0.6 mV in amplitude. The α-particle spectrum showed a FWHM of 29%. An important observation is that the crystal did not show any tendency towards polarization at this temperature, i.e., no pulses were observed when the applied electric field was switched to zero while the crystal was being irradiated. This particular feature can be very advantageous where the crystal is to be used as a counter over extended periods of time and frequent optical illumination is not practicable. A specific application is in the detection of γ ray or electron showers at high energies where the high atomic number of this material can enable the experimenter to design total absorption shower detectors of reasonably small volume and 100% efficiency.

KRS-5

This is a eutectic mixture of TlBr and TlI. A few crystals of this material having an impurity content less than 10 ppm were studied as described above. However, the α-particle experiment we carried out showed conclusively that only electrons were mobile in these crystals. Hole movement, if present, was too small to be observed, i.e., hole pulses, if any, were below the 0.2-mV noise level of the preamplifier. The energy per ion pair in KRS-5 measured with the γ rays of ^{60}Co turns out to be 18 ± 2 eV. We estimate the value of Ψ to be 0.1. γ pulse-height spectra, Figs. 8(a) and 8(b), do not show any structure at all, and this is explained by the simple reason that only one type of carrier is responsible for the conduction pulses. The α-particle spectrum, Fig. 8(c), shows a spread of about 50%. No sharp optimum temperature could be found for this material. There is a wide range of temperatures extending from -172 to $-117\,^{\circ}$C over which the response of this crystal is sensibly flat. The efficiency for ^{60}Co γ rays was measured to have the value of about 10%. The α-particle efficiency was close to 10%. The dielectric constant of KRS-5 over the useful range of temperatures was measured to be 37.

THALLIUM CHLORIDE

Pulses of up to 2-mV amplitude were counted with a crystal of this material on bombarding it with a ^{60}Co γ-ray source at an electric field of 2100 V/cm and a temperature of $-160\,^{\circ}$C. This included the factor of 60 amplification of the charge-sensitive preamplifier. The energy per ion pair calculated from the observed pulse height from ^{60}Co γ rays turned out to be 52 eV. The crystal could be cleared of the accumulated space charge by shining white light on it as described earlier. The value of Ψ, by assuming the optical band gap to be 4000 Å or 3.1 eV, is 0.06. The crystal resistance at $-180\,^{\circ}$C was nearly 10^{12} Ω. The crystal also showed photoconductivity similar to other thallium halides we studied. A polarization curve for this crystal is shown in Fig. 9. This figure shows the typical way in which the counting rate decreases with time when the crystal is counting γ rays. The efficiency for counting γ rays from ^{60}Co was nearly the same as that of TlBr. The dielectric constant of TlCl was measured and found to be nearly 33.

SPACE-CHARGE EFFECTS

All thallium halide crystals studied were found to be subject, at low temperatures, to polarization due to space charge created by the trapping of moving electrons and/or holes. In all cases the space-charge field built up much more quickly when the crystal was bombarded by α particles and more slowly when bombarded by γ rays.[17] Before taking any data, any built-in field was removed by shining white light in the case of TlBr and TlCl and red light (white light through a sharp cutoff CS-2-58 optical filter) in the case of KRS-5. The disappearance of the space charge was indicated by the gradual dwindling of the pulses that are observable even at no applied bias under bombardment by any of the radioactive sources

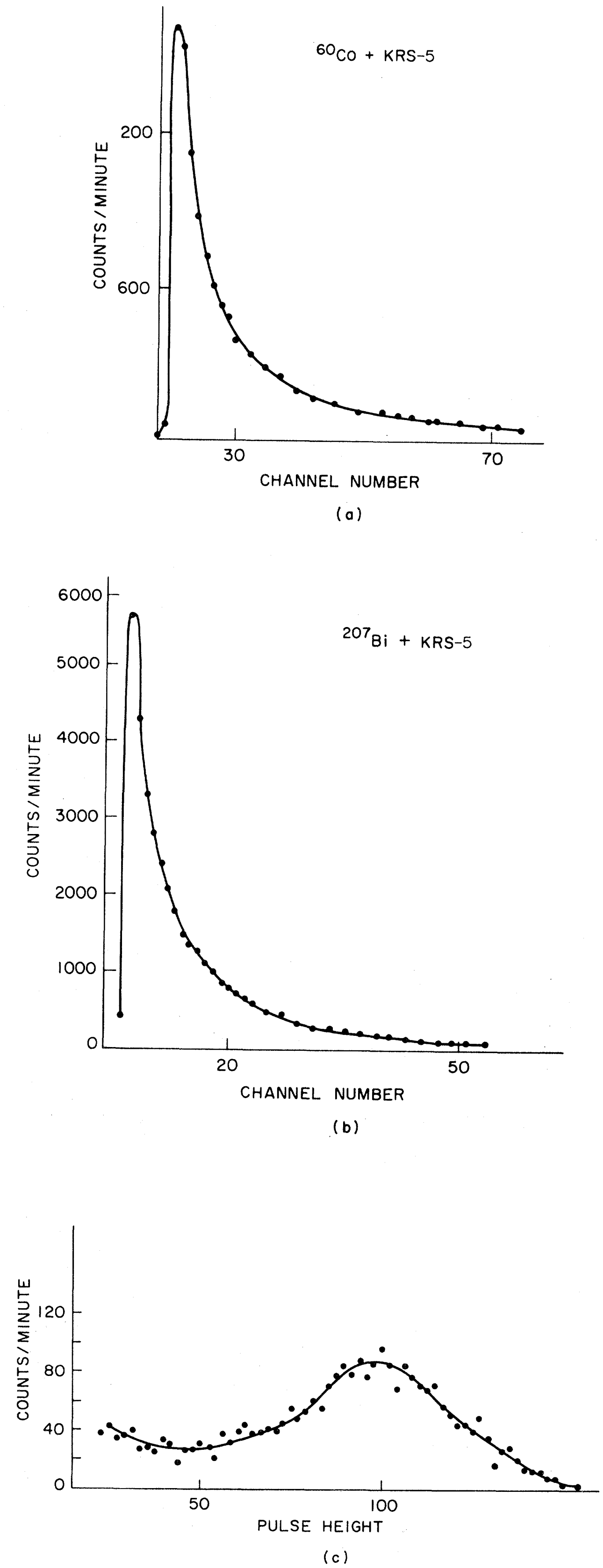

FIG. 8. (a) Pulse-height spectrum due to γ rays from ^{207}Bi obtained with a KRS-5 conduction counter. (b) Pulse-height spectrum due to γ rays from ^{60}Co. (c) Pulse-height spectrum due to α particles from ^{241}Am.

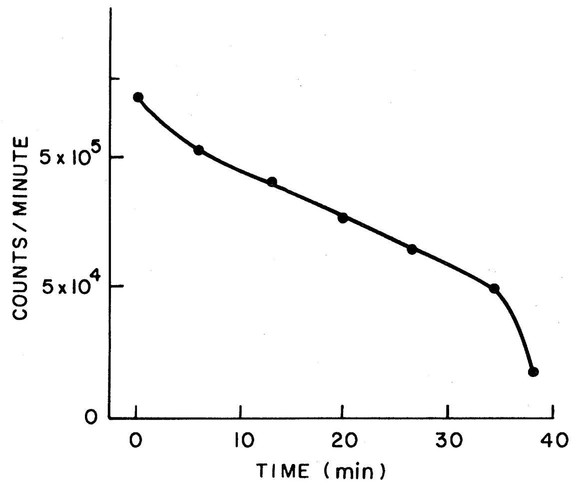

FIG. 9. Variation of the counting rate for γ rays with time due to polarization in a TlCl conduction counter. For details see text.

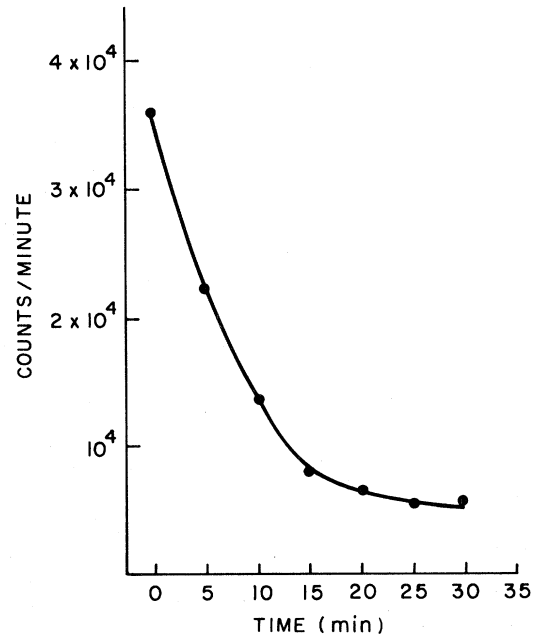

FIG. 10. Variation of the counting rate for α particles as a function of time due to polarization in a TlBr conduction counter.

used. The time required to effect this "cleaning" procedure was of the order of a few seconds. The space-charge buildup caused the size of the pulses to diminish with time; thus the pulse-height spectra were taken over a short period of time of 20—60 sec in the case of α particles and 2—5 min in the case of γ rays. A typical counting rate versus time curve for α particles is shown in Fig. 10.

CONCLUSION

It should be noted that our results have been obtained on a small number of crystals and in the case of TlBr, only one sample. We are in process of obtaining additional crystals for our studies. Our work on the counting properties of various thallium halide crystals has shown clearly that in a "pure" TlBr crystal both holes and electrons can move in comparable proportions at a temperature of approximately $-90\,^{\circ}\mathrm{C}$. The energy resolution achieved with monoenergetic α particles is moderately good and is comparable with that obtained with a crude CsI(Tl) scintillation counter. The efficiency for detecting α particles is close to 100%. These crystals of high atomic number and high-density materials can detect γ rays with fairly good efficiency, and the limit is probably set only by the size of the crystal. Thus these materials, particularly TlBr, may become useful detectors for high-energy physics, since they may act as total absorption shower counters.

The electron and hole contributions to conduction pulses depend strongly on temperature in TlBr. No such effect has been observed in KRS-5, whose response is sensibly flat over a wide range of temperatures. Surface effects and inhomogeneities or strains in the body of the crystal may be responsible for much of the observed experimental resolution for α particles. On the other hand, crystals in which only one type of carrier gives conduction pulses have poor resolution for γ rays although the resolution for α particles still depends on the factors stated above.

The smaller electronic contribution in TlBr and the unobserved hole movement in KRS-5 can be caused by the existence of deep-lying electron and hole traps, respectively. A study of the trap depths may throw some light on the observed behavior of these conduction counters.

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tive electrode, the resulting pulse will be $e[x/d + (d-x)/d] = e$. If all carriers correspond to this situation the resulting pulse distribution will be homogeneous.

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