Behavior of Space Charge in Diamond Crystal Counters under Illumination. I*

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The development of a technique is described whereby alternate electric field pulses and white light flashes are used to overcome the difficulty of the formation of space charge in a diamond crystal counter when subjected to bombardment by β-particles. Using the method, a study is made of the behavior of the space charge field when the electric field strength and flash intensity are varied. The manner in which the space charge grows and decays with time depending on the conditions of the experiment is described. It is shown how, under suitable conditions, steady counting-rates can be obtained and in particular, how the behavior of the counter under space charge free conditions can be investigated. Qualitative results are also given concerning the effect of illumination of the counter with red and infrared light instead of white light.

Preliminary experiments on the effect of temperature on the counter suggest the possibility of permanently preventing the formation of space charge by keeping the temperature of the counter sufficiently high. It is also indicated how experiments at different temperatures could yield information concerning the distribution of trap depths in the crystal and the mean lifetimes of trapped electrons and holes.

A new approach to the problems of solid state physics has thus been developed. The great advantage that it possesses over luminescence methods is that it does not require the emission of light in order to study the electron transitions between trapping states and the ground state and consequently, radiationless transitions can be investigated.

A FUNDAMENTAL disadvantage that as yet limits the use of conductivity crystal counters as detectors for nuclear radiations is the continual growth of a space charge field inside the crystal as the bombardment proceeds. This space charge field opposes the applied field, thus causing the height of the charge pulses produced by particles of given energy to decrease with time. Such behavior is obviously undesirable if the counter is to be of practical use; and, accordingly, a technique has to be devised whereby this difficulty can be overcome. Methods which allow the behavior of the crystal to be studied under conditions of freedom from space charge have been described by several authors. One of the earliest methods was that used by Gudden and Pohl[1] for studying the photoconductivity of diamond. When a space charge had formed, it was removed during a period in which the crystal was illuminated with red light alone, thereby causing the trapped electrons and holes to be released. Another method that has been used is to apply alternating fields to the crystal;[2] the space charge that forms during the first half-cycle is then neutralized by an equal but opposite space charge field formed during the second half-cycle. Recently, this technique has been used very successfully by McKay;[3] the applied field followed a sine wave and the bombardment with a high intensity beam of low energy electrons took place at only the positive and negative crests of the field cycle. This arrangement, though admirable for the purpose of the above experiments in which the actual response of the crystal to radiations was being studied, would not be so suitable when using the crystal as a counter, since the crystal would be inactive for much the greater part of the field cycle. By making the applied field of square-wave form this criticism could be to a large extent removed, and in experiments where the counter field could be synchronized with some other function of the apparatus, as in a pulsed output from a cyclotron, this arrangement would possibly prove quite suitable.[2]

Summarizing, although the space charge difficulty can be overcome in certain cases, none of the above methods are entirely satisfactory if the crystal is to be used as a counter for nuclear experiments on account of the relatively long intervening periods required for depolarizing the crystal. In this case, it is much more desirable to have a continuously sensitive counter, or at least a counter that is operative for a large proportion of the time of the experiment.

PRELIMINARY EXPERIMENTS WITH ILLUMINATION

In diamond the space charge field that opposes the applied field is caused by electrons and positive holes that have become held in trapping centers throughout the crystal. Little is known about the nature of these traps in diamond, especially those for the positive holes; but for electrons, it has been reported[4,5] that their depths appear to lie between 0.25 and 0.75 electron-volt. It should therefore be possible to eject these trapped electrons by irradiating the crystal with light of appropriate wavelengths, i.e., with the red and near infrared part of the spectrum. The ejection of trapped electrons in this way, using light or by warming the crystal, has been suggested before,[6] although few detailed reports of the use of light in this respect, i.e., in connection with crystal counters, have been published. In an earlier letter[7] the author reported the

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[1] B. Gudden and R. Pohl, Z. Physik **16**, 170 (1923), *et seq.*

[2] L. F. Wouters and R. S. Christian, Phys. Rev. **72**, 1127 (1947).

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

[4] R. R. Newton, Phys. Rev. **75**, 234 (1949), quotes the experiments of A. J. Ahearn to estimate that the trap depths lie between 0.25 and 0.75 electron-volt.

[5] C. Bull and G. F. J. Garlick, Proc. Phys. Soc. (London) **A63**, 1283 (1950).

[6] E.g., R. Hofstadter, Nucleonics **4**, 14 (1949).

[7] A. G. Chynoweth, Phys. Rev. **76**, 310 (1949).

maintenance of a steady counting rate in diamond crystal counters under alpha-ray bombardment by simultaneously illuminating the crystal with the light from a Nernst filament. These results showed that the illumination was efficient in removing the space charge field that formed in the crystal, although their complete interpretation must take into account some ionization of the surrounding air by the α-particles. Because of this effect, quantitative examination of the results was not possible. Results which seem to confirm, at least partially, the author's findings have been given by Freeman and van der Velden.[8] Willardson and Danielson[9] have also published somewhat similar results of experiments in which diamond crystal counters were illuminated while under bombardment with beta-rays.

In repeating the experiments of Willardson and Danielson, the author found several diamonds that responded to the beta-rays from radium E; but, when illuminated with the unfiltered light from the Nernst filament, they all showed behavior very similar to that of the specimen TSC-2 of Willardson and Danielson; that is, no great or permanent improvement in the counting rate was obtained on illumination. At its commencement, however, the counting rate did exhibit a transient increase, this increase quickly giving way to a continuing decay. The rate of decay under illumination was found to increase with the intensity of the light. A similar behavior was obtained with either of two arrangements of the electrodes: (i) when placed on opposite parallel faces of the crystal, (ii) when on the same face and separated by a gap of about 2 mm. The explanation of these phenomena is possibly as follows. In the dark, as the bombardment proceeds, a space charge will accumulate, the electrons being trapped mainly towards the anode and the positive holes towards the cathode. In the given experimental conditions it is likely that, except at the lowest applied fields, the "centers of gravity" of these negative and positive space charge distributions will be fairly close to their respective electrodes. In this case, it is the net field (equal to the difference between the applied field and the space charge field) which will be the main factor controlling the size of the pulse caused by a particle of given energy, especially with the second electrode arrangement. The effect of illuminating the crystal with visible and infrared wavelengths will be to cause many of the trapped electrons and holes to be freed. Under the action of the field they can then move closer to the electrodes, either reaching them or becoming retrapped on the way. Hence, the over-all effect will be that the centers of gravity will have moved apart to a slight extent and, therefore, the net field over the greater portion of the crystal lying between the planes parallel to the electrodes and passing through the centers of gravity will have increased slightly. Consequently, there will be a small rise in the pulse height and, likewise, the counting rate will show an increase, since there is a wide distribution of pulse heights. Thereafter, as the bombardment continues, the space charge will proceed to accumulate as before, though in its new distribution, and again the counting rate will decay. If the light is itself capable of creating electron-hole pairs by some mechanism, either from the lattice atoms, from any impurity atoms present, or by any other process, the space charge will grow still more rapidly and the counting rate will show a steeper rate of decay.

Since all the available crystals that responded to beta-rays showed a similar behavior to that just described, illumination of the crystal simultaneously with the bombardment was useless as a means of removing the space charge difficulty; in fact, the difficulty was made worse by the illumination. Consequently, a reversion had to be made to pulsing techniques in order to study the space charge free behavior of the crystals. In order that the inoperative time of the counter could be reduced to a minimum, the scheme that was finally adopted was as follows. Alternately, a field pulse and a light flash were applied to the crystal. During the field pulse, with the crystal in the dark, a space charge formed. The field was then removed and a brief light flash was given to the crystal. The light caused many of the trapped holes and electrons to be released; and, when free, they could drift through the crystal lattice under the influence of the residual space charge field. The latter would cause the electrons and holes to drift towards each other, and from the results of McKay's work it appears probable that most of these freed electrons and holes would recombine, thus reducing the space charge field. Even if recombination were far from complete, the electrons and holes would have drifted nearer to each other (i.e., the positive and negative space charge regions would have intermingled), and the net effect of the light flash would again be a reduction in the space charge field. In addition, the light may also free electrons and holes from the lattice and impurity atoms. When free, these would behave just like those electrons and holes freed from traps and the space charge neutralization process would take place even more rapidly. If, therefore, the light intensity were sufficiently great, almost all the space charge formed during the field pulse could be dispersed and neutralized during the subsequent light flash. Hence, when these conditions had been reached, any further increase in the light intensity produced no appreciable change in the recorded value of the counting rate. With increasing light intensity the neutralization process could be completed more rapidly, thus making it possible to operate the counter in the normal manner for a greater proportion of the time. With very high light intensities, as long as the rate of influx of charged

[8] H. A. van der Velden and G. P. Freeman, Physica **16**, 493 (1950). Also G. P. Freeman and H. A. van der Velden, Physica **16**, 486 (1950).

[9] R. K. Willardson and G. C. Danielson, Phys. Rev. **77**, 300 (1950).

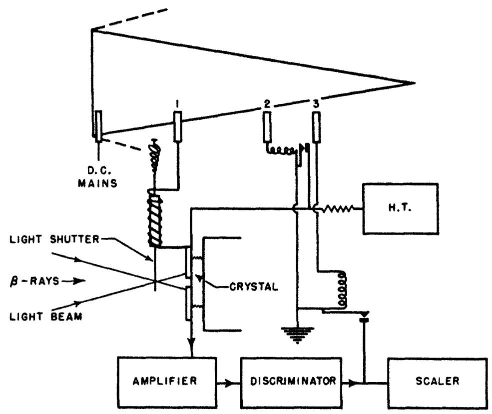

FIG. 1. Schematic representation of the experimental arrangement.

particles was not too great, the duration of the field pulse could be made much greater than that of the light flash.

EXPERIMENTAL ARRANGEMENT

The experiments to be described below were all made using the second arrangement of the electrodes, i.e., the two electrodes were placed on the same face of the crystal. The brass electrodes were made thick enough to prevent the most energetic of the beta-particles used from reaching the crystal except through the gap between them. Experiments were made in which (i) the brass electrodes which were clamped on to the diamond made contact with it via evaporated aluminum electrodes also separated by the same distance, and (ii) no evaporated electrodes were used. It was found that the behavior of the counter was similar, qualitatively, in both arrangements and, in most of the experiments described, no evaporated electrodes were used. The fact that the behavior was very much the same in both these arrangements suggests that, as expected, the majority of the trapping centers for the electrons and holes responsible for the accumulation of a space charge at the low applied fields used exist in the body of the crystal and not at the crystal electrode boundaries.

Though preliminary tests were made on a number of diamonds, all the experiments to be described were performed on only one specimen. The diamond used was a twinned triangularly shaped crystal about three mm thick. On macroscopic examination, it appeared to be very free from mechanical flaws such as laminations and fractures. Between crossed polaroids, most of the crystal was seen to be strongly birefringent except at the vertices of the triangle, and there were two dark streaks running right across the crystal from two of the vertices to the opposite sides. The ultraviolet transmission spectrum of the diamond showed that it transmitted down to wavelengths of at least 2500A.

The crystal and the electrodes were assembled on a polythene insulator in such a way that no polythene was actually exposed to the radiations. One electrode was attached to the control grid of the input valve by a very short wire, the grid leak being 3×10^9 ohms. The other electrode could be held at any positive or negative voltage required. A slow, non-feedback amplifier of the type used for electron collection in gas ionization chambers was employed; it had time constants of the order of 10^{-4} sec. The output pulses from the amplifier were fed into a scaling circuit via a pulse height discriminator. To keep the temperature of the crystal steady, a heating coil was placed around the crystal holder and a thermojunction for recording the temperature was mounted so that it almost touched the crystal.

The light from the Nernst filament was focused on to the crystal by an aluminized concave mirror and the flashes were controlled by a light shutter that was caused to move sharply in and out of the light beam by electromagnets. Light intensities were measured with a photoelectric cell. The field applied to the crystal could be removed by earthing the H.T. line through a leak resistance using an electromagnetic relay. Hence, the field pulse and the light flash could be alternated by suitable timing of the relay and electromagnetic circuits; this latter was controlled by a commutator, the action of which can be understood by referring to Fig. 1. The commutator consisted of a cylindrical drum of Tufnol, one foot long and three inches in diameter. It was rotated at a constant speed of 30 rpm by a synchronous motor. Wrapped round this drum and stretching from one end to the other was a thin sheet of soft copper, which if flattened out, would have the shape of an isosceles triangle. The apex of the triangle was clamped at one end of the drum, while the base was wrapped round the other so that the base angles of the triangle just overlapped. Pressing against the drum as it rotated were four phosphor bronze contacts, one of which was in continual contact with the base end of the triangle. Through the latter current was fed from the dc mains; and, when the triangular sheet completed the circuit, the current was tapped off via the other contacts to activate the electromagnetic relays.

As the drum rotated, contact 1 was the first to be made, and this activated the electromagnet which caused the shutter to cut off the illumination from the crystal. Next, contact 2 was made causing the field to be applied to the crystal. Disregarding for the moment contact 3, it is seen that as the rotation continued, contact 2 was the first to be broken, thereby cutting off the field. This was followed by the breaking of contact 1 which caused the electromagnet to withdraw the shutter from the light beam, thereby illuminating the crystal. By suitable placing of the contacts 1 and 2 along the drum, the ratio of the durations of the field pulse and the light flash could be varied within very wide limits. In practice, it was found that sparks at the various makes and breaks resulted in an undesirable background counting rate even when a large number of smoothing circuits had been added. Consequently, a third electrode was placed on the drum as shown. This activated another relay immediately after the field had been switched on, causing the pulses from the discriminator to be fed into the scaler. When contact 3 was broken immediately before removing the field, the pulses were diverted to earth. Hence the scaler was operative only while a field was applied to the crystal, and during this time no other makes or breaks occurred. In this way the background counting rate due to all causes (radioactive contamination, spurious electronic pulses, and the pick-up of external transients) was found to be reduced to the order of, at most, 5/minute and was therefore entirely negligible compared with the counting rates used.

FIELD-LIGHT CYCLE

During the field pulse, the space charge will show an increase, this being followed by a decrease during the subsequent light flash. Let the space charge field be represented by the number of trapped electrons, n, and let N_0 correspond to the space charge field that exactly balances the applied field. Suppose that at the commencement of a light flash there are N_0 trapped electrons. Then, during the flash this number will decrease to a value N_1, only to increase again to a value N_1' during the subsequent field pulse. If the intensity of the light, I, is sufficiently great, N_0-N_1 is greater than $N_1'-N_1$. The space charge field will therefore vary with time in a manner similar to that shown in Fig. 2; and, in time, a condition of equilibrium will be attained in which the amount of space charge formed during a field pulse is equal to the amount removed during a light flash.

During the light flash, the fall in the space charge may be ascribed to several causes:

1. The light will release trapped electrons and holes, the probability of an electron being released in time dt being proportional to $nIdt$.

2. Over the region into which the beta-particles penetrate, they also (and probably the freed electrons as well) will be able to release trapped electrons. The probability of the release of an electron by this method in time dt will be approximately proportional to $npdt$, where p is the intensity of the beta-rays.

3. It is quite possible that the light can liberate electrons directly from the lattice atoms, though it is known that, in diamond, the quantum efficiency of this process is very low.[10] Furthermore, only relatively few of the quanta present in the incident spectrum are capable, theoretically, of releasing lattice electrons. The contribution of this process to the decay of the space charge is difficult to formulate although a first approximation may be made as follows. The contribution to the decay of the space charge by this method will be zero when the space charge field is zero (no net field during light pulse to separate appreciably the newly formed electrons and holes). The effect will be a maximum when the space charge field is a maximum. Accordingly, suppose that, to a first approximation, the probability of an electron being released from the lattice to set up an opposing space charge is proportional to the space charge field, i.e., n. Therefore, the probability per unit time of an electron being freed by this process is proportional to nI.

4. The beta-rays will also liberate lattice electrons, thus causing a decay in the space charge field in the same way as the light, and therefore the probability per unit time of an electron being freed by this process is, to a rough approximation, proportional to np.

5. Finally, electrons and holes will be released from their traps thermally. McKay[3] describes experiments that show that at room temperatures, the rate of thermal ejection of at least some of the electrons is very rapid (half-life of the order of microseconds) through the positive holes are released relatively slowly. If all the electrons were released rapidly, then they would either recombine with the holes or distribute themselves in such a way as to neutralize the space charge of the holes. However, if a space charge is formed inside the diamond, which is then left in the dark without an applied field, a residual space charge can still be detected on bringing up a β-ray source, even after several hours have elapsed. Recently, Bull and Garlick[5] have described experiments in which glow curves were obtained with diamonds left for several hours at room temperature in the dark after being activated. It therefore seems likely that there are two active types of electron trap, i.e., one in which electrons have a very short lifetime at normal temperatures and the other showing a very slow rate of release (half-life of the order of minutes, or hours). For the present purposes, we can assume that the electrons that are trapped in the short-life traps are released in a time that is very short compared with the lengths of the light flash and the field pulse, and hence do not play any part in the space charge fields that are being considered.

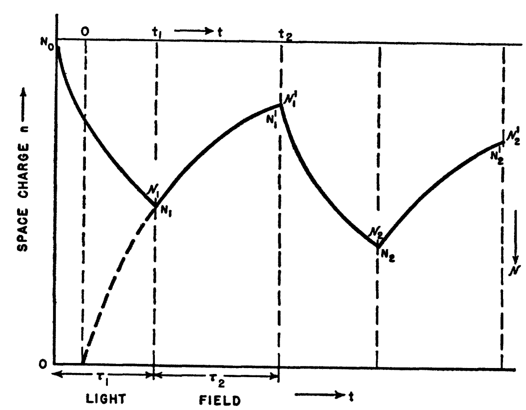

FIG. 2. Representation of the variation with time of the space charge field during the field-light cycle.

[10] See N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, New York, 1948), p. 140.

The appropriate differential equation approximately representing the decay of the space charge during a light flash will therefore be

$$dn/dt=-anI-bnI-cnp-dnp, \tag{1}$$

where the first and second terms represent the release of trapped and lattice electrons, respectively, by light, the third and fourth terms represent the release of trapped and lattice electrons, respectively, by the beta-rays, and a, b, c, d are constants.

Experimentally, it was found that when the crystal was illuminated by light of any except the lowest intensities used, the space charge decayed much more rapidly than when the crystal was subjected to the beta-ray bombardment alone. This indicated the greater effectiveness of the light in removing the space charge. Consequently, save for the lowest values of I, only the first two terms of Eq. (1) are of importance and it then becomes

$$dn/dt=-\alpha In, \tag{2}$$

where $\alpha=(a+b)$. Imposing the condition that $n=N_0$ when $t=0$, the solution of this equation can be written:

$$n=N_0\exp(-\alpha It). \tag{3}$$

The growth of the space charge during a field pulse is caused by the formation of free electrons and holes by the beta-rays. Normally, the crystal is in the dark during a field pulse but the effect of illuminating the crystal simultaneously with the application of the field will also be considered, the light assisting in the formation of space charge by liberating lattice electrons and holes. Again, the light and beta-rays will be capable of releasing trapped electrons and holes; but, owing to the applied field, these will be prevented from recombining with each other. These electrons and holes which were originally trapped will move towards their respective electrodes probably becoming re-trapped, and hence the net space charge due to this effect alone will not alter greatly.

As the total number of traps which can be occupied is N_0, the number of unoccupied traps at a given instant is effectively N_0-n. Therefore, the probability of a free electron being trapped in time dt is proportional to $(N_0-n)dt$ as long as all the freed electrons become trapped within the crystal. It will be assumed that, with most of the field strengths used in the experiments, this condition was obeyed approximately. Next, the probability of a lattice electron being released by the beta-rays in time dt is proportional to p, and therefore the probability per unit time that a beta-ray may form a space charge electron is proportional to $p(N_0-n)$; and similarly, for the light, it is proportional to $I(N_i-n)$. The differential equation representing approximately the growth of the space charge can then be written:

$$dn/dt=a'p(N_0-n)+b'I(N_0-n), \tag{4}$$

where a' and b' are constants. Since $n=0$ when $t=0$, the solution to this equation is

$$n=N_0[1-\exp(-\beta t)], \tag{5}$$

where $\beta=(a'p+b'I)$. For convenience a change of coordinates will be made; i.e., when n traps are occupied, let $\mathfrak{N}$ operative traps be unoccupied. Then $n=N_0-\mathfrak{N}$ and substituting into Eq. (5):

$$\mathfrak{N}=N_0\exp(-\beta t). \tag{6}$$

Equations (3) and (6), though very much over-simplified, will give an approximation to the behavior of the space charge during the light field cycle; and, in particular, they describe the effect of the intensity of the light and the ultimate steady conditions. We proceed as follows. During the first light flash, the space charge decays from N_0 to a value N_1, where, from (3),

$$\left.\begin{aligned} &N_1/N_0=\exp(-\alpha I\tau_1)=A \text{ (say)},\\ \text{i.e.,}\quad &N_1=AN_0,\end{aligned}\right\} \tag{7}$$

where τ_1 is the duration of the light flash. The quantity A depends only on the intensity of the light [assuming that the approximate Eq. (2) can be used instead of (1)]. The space charge then grows during the following field pulse to a value N_1', after which it decays to a value N_2, where

$$N_2=AN_1'. \tag{8}$$

Similarly, $N_3=AN_2'$, etc. Also, if at the end of the first light flash the number of unoccupied traps is $\mathfrak{N}_1$, this number decreases to $\mathfrak{N}_1'$ during the subsequent field pulse where, from (6),

$$\mathfrak{N}_1'/\mathfrak{N}_1=\exp(-\beta\tau_2)=B \text{ (say)}, \tag{9}$$

where τ_2 is the duration of the field pulse. B depends only on the intensity of the beta-ray bombardment, since there is no illumination during the field pulse, i.e., $I=0$. Then,

$$\mathfrak{N}_1'=B\mathfrak{N}_1. \tag{10}$$

Similarly, $\mathfrak{N}_2'=B\mathfrak{N}_2$, etc. Using the above relations the values of N_1, N_2, $N_3\cdots$ in terms of N_0, A and B can be determined. In particular, it can easily be shown that the value of the space charge remaining after r light pulses is given by

$$N_r=AN_0\left[1-\frac{(B-AB)(1-A^{r-1}B^{r-1})}{(1-AB)}\right]. \tag{11}$$

Since r can be taken to represent the time that has elapsed since the commencement of the cycles, this equation gives the way in which the residual space charge approaches its steady value. To determine the ultimate residual space charge, N_∞, let r tend towards infinity and then, since A, B are both less than unity,

$$N_\infty=AN_0(1-B)/(1-AB). \tag{12}$$

The findings of this section will now be compared with experiment.

EXPERIMENTAL RESULTS FOR THE GROWTH AND DECAY OF THE SPACE CHARGE

From Eqs. (3) and (6) it is seen that the space charge should decay and grow approximately obeying exponential laws. This deduction was tested experimentally by the following technique. Using initially a high light intensity during the field-light cycle, a curve showing the variation of the counting rate with the applied field was obtained, i.e., under virtually space charge free conditions. Next, a saturation space charge was formed in the crystal (by the action of light and beta-rays); by "saturation" is meant that the space charge field was equal in magnitude to the applied field. The time required for this condition to be obtained was determined by separate experiment. On removing the field, space charge pulses were obtained, the size and counting rate of these decreasing steadily with time as the space charge field became neutralized. The counting rate decay curves were obtained by noting the times of occurrence of each successive hundred pulses recorded by the scaler enabling an integral curve (total number of pulses recorded with time) to be plotted. Differentiating this gave the counting rate decay curve for the space charge pulses. Cross interpolating between this curve and that of counting rate against field, it was possible to deduce the decay of the net field inside the crystal with time. These curves were obtained when the crystal was in the dark (the beta-rays then being the only agent neutralizing the space charge) and when illuminated at various intensities. The results are shown in Fig. 3, where the logarithm of the space charge field is plotted against time. From Eq. (3) this should yield a straight line and it is seen that those curves obtained using the higher light intensities are indeed satisfactorily linear. At the lower light intensities and in the dark, the curves show marked deviations from the straight line form. These deviations are probably due to the action of the β-rays and light in releasing lattice electrons, for it is very unlikely that the simple relations that were used in the above theory are very satisfactory. In particular, when the space charge field is very low, it is likely that the probability of a newly formed electron-hole pair separating from each other to add to the space charge field instead of recombining is very much higher than that given by assuming it to be proportional to the net field in the crystal. This effect in itself could probably explain the deviations observed. The fact that the deviations from the straight line are not nearly so apparent at the higher light intensities seems to indicate the far greater effectiveness of the β-rays in freeing lattice electrons. No attempt has yet been made to try other functions for the probability of release of a lattice electron, as it is felt that the accuracy of the curves does not warrant such a procedure. More recent modifications to the apparatus may lead to information concerning the various processes involved in the release of the lattice electrons.

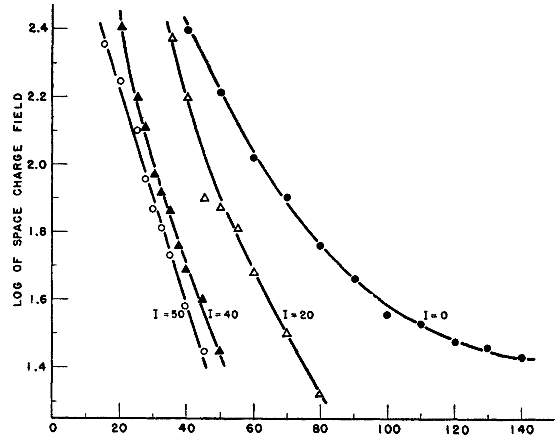

FIG. 3. Log plot of the decay of the space charge field with time for various intensities of illumination of the crystal. The intensity, I, is in arbitrary units.

Besides the errors in the theory, there are more errors inherent in the experimental procedure. They are:

1. Examination of the decay curves shows that when the net field inside the crystal is high or low, the counting rate for the space charge pulses changes slowly. This makes accurate differentiation of the integral curves more difficult over these regions with a corresponding loss of accuracy in the space charge field decay curves. For medium fields, however, satisfactory accuracy is obtained.
2. Slight errors in the counting rate as obtained by differentiating the integral curves over the region corresponding to a high net field result in relatively large errors in interpolating the net field from the curve of counting rate against field as the latter approaches saturation at high fields.
3. At low counting rates, i.e., at low fields, the counting rate-field curve is not so accurate as at higher fields, since the recording of a smaller total number of pulses at these fields introduces a relatively larger random counting error.
4. It has been assumed that the counting rate-field curve was obtained under conditions of freedom from space charge. Although relatively high light intensities were used, there must still have been a small residual space charge which would have a slight effect on the shape of the curve.

The general conclusions are therefore that the experimental curves are satisfactorily accurate except at very high and low net fields in the crystal. At low light intensities, the dominant process causing the neutralization of the space charge must presumably be the liberation of lattice electrons by the beta-rays; and it must be concluded that, as expected, this process does not obey such a simple law as that indicated. There is no reason to doubt that the process of liberating the lattice electrons by the light must also be more complex, neither of these processes resulting in the simple expo-

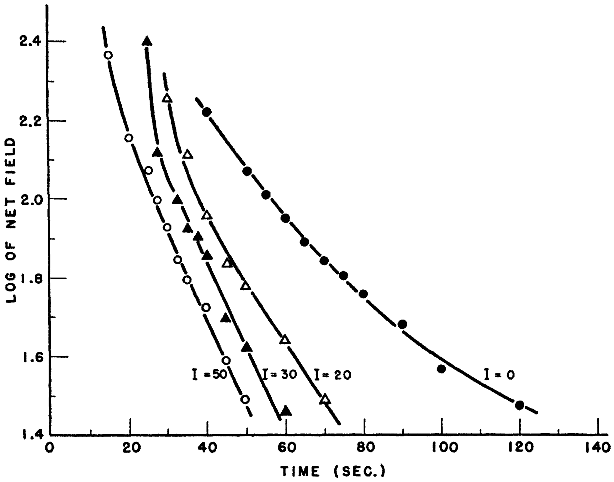

FIG. 4. Log plot of the decay of the net field with time for various intensities of illumination. The intensity I, is in arbitrary units.

nential decay law. However, at the high light intensities, the simple law is obeyed (to within the limits of the experiments), and the conclusion is that under these conditions the dominant process is the liberation of trapped electrons by the light as opposed to the liberation of lattice electrons. In particular, in most experiments the light intensity during the field-light cycle was well above 50 units (the highest values used in obtaining the decay curves); and, in fact, in obtaining the counting rate-field curves, it was of the order of 500 units. In this case it is probably sufficiently accurate to use the simplified Eq. (3) in the subsequent theory.

The curves showing the growth of the space charge under different intensities of illumination are shown in Fig. 4. (Actually, the log of the net field is plotted against time, and from Eq. (6) this should be a straight line.) They were obtained in a similar manner to those of Fig. 3, the crystal first being made free from space charge by illuminating it for a few minutes in the absence of a field. On applying the field the decay curves for the pulses were obtained. Since the growth of space charge is caused by the release of lattice electrons by the beta-rays and the light, it is not expected in view of the foregoing paragraphs that the log plots of the space charge field with time will be perfectly linear. It is again evident from Fig. 4 that a more complex law than that assumed in the above theory is necessary; however, Eq. (6) should enable a qualitative prediction regarding the behavior of the space charge in the field-light cycle.

TABLE I. Value of the constant, B, as obtained over different regions of the curve showing the growth of space charge with time under the conditions of the experiment.

Range (sec)	B	Range (sec)	B
10–15	0.990	55– 60	0.953
15–20	0.973	60– 65	0.956
20–25	0.952	65– 70	0.961
25–30	0.948	70– 75	0.955
30–35	0.958	75– 80	0.954
35–40	0.960	80– 85	0.950
40–45	0.954	85– 90	0.950
45–50	0.959	90– 95	0.955
50–55	0.953	95–100	0.962

DEPENDENCE OF COUNTING RATE ON LIGHT INTENSITY DURING CYCLE

When equilibrium in the cycle has been attained, the space charge present is given by Eq. (12). If F_∞ is the net field inside the crystal corresponding to a value N_∞ for the residual space charge, then

$$F_\infty/F_0=(N_0-N_\infty)/N_0=1-(N_\infty/N_0),$$

where F_0 is the applied field (F_0 corresponds in magnitude to N_0). By means of this relation the net field existing inside the crystal under steady conditions can be determined in terms of A and B. If the value of B is known, arbitrary values for A can be substituted into Eq. (12) thereby giving values for the fraction F_∞/F_0. Since F_0 is known, F_∞ is therefore determined, i.e., the net field that corresponds to the value chosen for the constant A, and therefore depends only on the light intensity (in arbitrary units). Since the curve showing the dependence of the counting rate on the net field inside the crystal is known, the theoretical curve showing the dependence of the counting rate on the light intensity can be interpolated. This can be compared with the curve obtained directly by experiment and thus serves as a check on the theory developed in the above section.

First, the method of determining the value of B will be described. The crystal was first freed from space charge, after which a curve was taken showing the growth of the space charge field with time under the conditions of the experiment for which the counting rate-light intensity curves were obtained. From Eq. (7) it is seen that B is the ratio between the ordinates for any two points on the space charge growth curve,the points being at an interval τ_2 apart and the ordinates being measured downwards from the asymptote to which the curve approaches. The value of τ_2 was 1.5 seconds, and it is obvious that the ratio of the ordinates separated by this interval would be difficult to obtain with any accuracy. Hence the ratio between the ordinates separated by an interval of five seconds was obtained and the value of B deduced by assuming that the curve was approximately straight over an interval of five seconds. By carrying out these measurements over various parts of the curve the average value of B could be obtained.

Table I gives the values of B obtained from various regions of the curve; and it is seen that, except immediately after the commencement of the curve, they are reasonably consistent. Ignoring the first two values, the average value of B becomes 0.955.

Substituting this value and suitable values for $A(0\leqslant A\leqslant+\infty)$ into Eq. (12) and carrying out the processes described above, it was possible to plot the theoretical curves of counting rate against light intensity for various applied fields. In Fig. 5 are shown such curves for applied voltages of 600 and 300. The experimentally obtained points are also shown. The theoretical curves have been fitted to the experimental points in the following manner. Both curves should correspond for infinite light intensity, and thus the ordinates of the theoretical curve were not altered. The abscissas of the theoretical curves were then multiplied by the factor required to make the curves fit as well as possible over the initial rise. It will be noticed that the fit obtained in this way is not very good for the higher light intensities; this corresponds to conditions where the crystal is largely free from space charge. The fact that the experimental points reach saturation more rapidly than the theoretical curves probably results from the approximations made in the theory for the growth of the space charge field. In Fig. 4 it is seen that the tendency is for the space charge field to grow rather less than it would if the simple exponential law were obeyed. This makes the equilibrium space charge field somewhat less than that given by Eq. (12); and, hence, the experimental counting rate-light intensity curve will be expected to deviate in the manner obtained.

EFFECT OF CONTINUOUS ILLUMINATION OF THE CRYSTAL

It has been stated that it was found to be necessary to use pulsing techniques in order to disperse the space charge in the crystal as opposed to continuous illumination of the crystal. As various authors have reported results using the latter method, it is of interest to give our findings in more detail. The crystal used by the author in his earlier communication was found to respond only to alpha-rays and not to beta-rays. Consequently, a different crystal had to be used for the present experiments. As stated above, constant illumination of the β-particle counting crystal with the full spectrum from the Nernst filament caused a space charge to be created by the light itself. Since the ultraviolet absorption limit was 2500A for this diamond, it might be thought that light of wavelengths longer than 4000A could safely be used without raising electrons from the full band up to the conduction band. This proposal was tested experimentally as follows. The crystal was first made free from space charge by illuminating it with white light in the absence of a field. The light was then passed through a filter with a sharp cut-off at 6900A; i.e., only the red and infrared light reached the crystal. During this illumination with red light, the field was applied though the beta-rays were not allowed to strike the crystal. Finally, the field and light were removed and the crystal was then bombarded by the beta-rays. It was found that space charge pulses were present. Hence, it could only be concluded that light of wavelength longer than 6900A was still sufficiently energetic to liberate electrons by some mechanism inside the crystal. This is apparently contrary to the results of Freeman and van der Velden,[8] who found that (using alpha-ray bombardment) their crystal could be illuminated with light of wavelength longer than 6900A simultaneously with the bombardment without any apparent disturbing effect. However, they observed that a filter cutting off at 6000A was not successful and this suggests that, as with many of the physical properties of diamond, the intrinsic cut-off values for this effect varies from specimen to specimen. Nevertheless, there still remains the problem of the origin of the electrons that are liberated by wavelengths greater than 6900A. The presence of impurity levels at a depth below the conduction band less than, or of the order of, the energy of the transmitted quanta, could possibly be the source of these electrons; or perhaps successive transitions between various intermediate energy levels are possible.

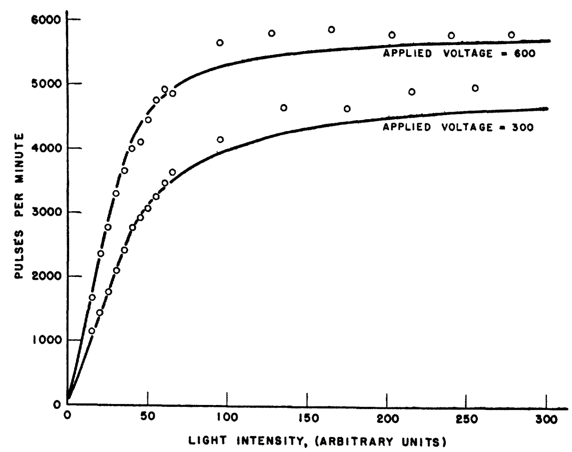

FIG. 5. Variation of the counting rate with the intensity of illumination during the field-light cycle. The full line shows the expected variation; the points plotted are those obtained experimentally.

In another experiment it was found that illumination of the diamond using the same filter in the absence of a field was still able to remove the space charge. This suggests that no, or very few, traps exist at an optical depth greater than that corresponding to 6900A, i.e., 1.8 ev. This conclusion is in agreement with the results of other workers[4,5] who have estimated the trap depths in diamond.

EFFECT OF TEMPERATURE ON THE SPACE CHARGE

The actual distribution of trap depths in diamond has been investigated[5] by obtaining thermoluminescence curves. These curves show that if the temperature of the crystal is raised to about 600°K, almost all of the electron traps become empty. In the glow curve there is a large peak at a temperature of about 520°K and a much smaller one at about 420°K. This indicates that most of the traps are at a thermal depth of about 520°K, which has been found experimentally from the

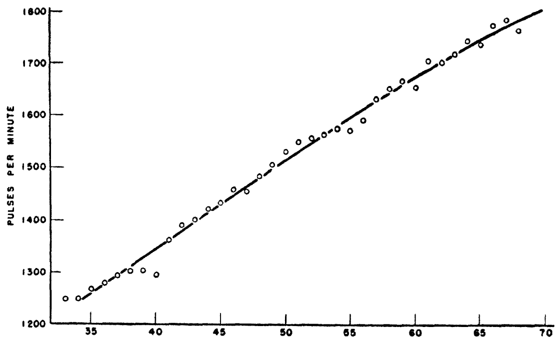

FIG. 6. Variation of the counting-rate with the temperature of the crystal. The warming and cooling rates were about 15°C per hour. Thirty-five divisions on the thermocouple scale approximately represent a temperature range of 30°C.

glow curves to correspond to roughly 0.7 ev. Hence, raising the temperature of the crystal above room temperature should enable some of these deeper traps to be thermally emptied and the net space charge thereby reduced. If the temperature were to be raised to 600°K, only a negligible amount of space charge should be left in the crystal, and it might be possible to obtain a continuously sensitive counter with a constant applied field.

The design of the apparatus did not permit raising the temperature by more than thirty or forty degrees above room temperature. However, over this range of temperature some traps could become empty, and hence the net space charge would be altered slightly. The apparatus was therefore arranged so as to detect any small change in the space charge, this being accomplished by applying a high field to the crystal and carrying out the usual cycle at a low light intensity. (When the cycle was continued without illumination, a certain amount of space charge formed during a field pulse was removed by the beta-rays in the subsequent "dark" pulse with the field removed. Empirically, it was estimated that this removal of space charge corresponded to an initial light intensity of 15 units. Consequently, in Fig. 5 the experimental points do not commence until the light intensity is 15. In particular, for the investigation into the effect of temperature, no illumination was used during the cycle.) From Fig. 5 it is then seen that if the residual space charge is reduced slightly (corresponding to a slight increase in the light intensity), the counting rate shows a relatively large increase.

Under the conditions just described, the temperature was raised steadily (at a rate of about 15°C/hour), the counting rate being recorded at regular intervals of temperature as recorded by the thermocouple. Having reached a sufficiently high temperature, the crystal was allowed to cool (at the same rate), still recording the counting rate.

The results are shown in Fig. 6. The graph was plotted by first taking, for any particular temperature, the mean of five successive readings about that temperature. This was carried out while the crystal was warming and again when cooling. The points plotted are the means of these two sets of readings. It is evident that the space charge was considerably altered by the rise in temperature, and for this reason it was important to keep the temperature of the crystal constant for all the experiments.

It seems very unlikely that this relatively large increase in the counting rate can be due to anything but a purely crystal phenomenon. To prove that it was not caused by spurious pulses due to the possible breaking down of the insulators at the higher temperatures, the beta-ray source was removed at the highest temperature attained and it was found that the background counting rate was less than 10/minute. A rough calculation showed that the increase in the counting rate caused by the increase in the noise level of the input circuit of the amplifier was also negligible, i.e., it was estimated that for a 30°C rise above room temperature, the maximum possible increase in the counting rate from this cause was 25/minute.

RETENTION OF SPACE CHARGE BY THE CRYSTAL

If the crystal were to be saturated with space charge and then left to itself, i.e., removing the beta-ray bombardment, the light irradiation, and the field, the only way in which the electrons could escape from the traps would be by thermal ejection. Considering traps of only one depth, E, the probability, p, of an electron escaping from a trap at a temperature T is of the form[11]

$$p = s\exp(-E/kT), \tag{13}$$

where k is Boltzman's constant, and s is a constant which may, however, vary slowly with temperature. (From the experiments on thermoluminescence,[5] s is found to be of the order of 10^6/sec in diamond.) If n is the number of trapped electrons at time t, the rate of escape of these electrons is then

$$dn/dt = -ns\exp(-E/kT). \tag{14}$$

For this formula to be correct it has to be assumed that there is negligible retrapping; i.e., a liberated electron combines directly with a positive hole under the action of the space charge field. If n is taken to be proportional to the space charge, then $n = N_0$ when $t = 0$, and the solution to Eq. (14) can be written,

$$\log_e n = -st\exp(-E/kT) + \log_e N_0, \tag{15}$$

and hence plotting $\log n$ against t should yield a value for $s\exp(-E/kT)$. Essentially this relation has been tested by Bull and Garlick using data from the glow curves; and it was found to hold reasonably well except for small values of t, i.e., t less than three hours at room temperature. This indicates that most of the traps in diamond can be regarded as being of the same depth;

[11] J. T. Randall and M. H. F. Wilkins, Proc. Roy. Soc. (London) **184A**, 366 (1945).

from the glow curves this depth is found to be approximately 0.7 ev.

In principle, the depths of the traps in diamond can also be obtained from measurements of the decay of the space charge. As described above, values for $s\exp(-E/kT)$ could be obtained over a range of temperatures. Let $f(T)=s\exp(-E/kT)$. Then we have, assuming that s remains fairly constant,

$$f(T_1)=s\exp(-E/kT_1) \quad \text{and} \quad f(T_2)=s\exp(-E/kT_2),$$

where T_1 and T_2 are any two temperatures. Hence,

$$\log_e\left[\frac{f(T_1)}{f(T_2)}\right]=\frac{E}{k}\left(\frac{1}{T_2}-\frac{1}{T_1}\right) \tag{16}$$

and therefore, from the experimentally obtained plot of $f(T)$ against T, E and finally s can be determined.

The techniques developed by the author allow relation (15) to be tested. It was desired to measure the space charge n still existing in the crystal after having been left undisturbed for a time t. To do this, the crystal was first saturated with space charge by irradiating it with intense light in the presence of a field. The light was then removed and the field switched off. After a time t had elapsed (t being anything up to several hours), the beta-ray bombardment was commenced and the initial slope of the integral decay curve gave the counting rate corresponding to the space charge field remaining in the crystal after a time t. From the curve of counting rate against applied field as obtained under space charge free conditions, the corresponding space charge field could then be interpolated. This process could be repeated for various values of t and hence a graph of the log of the space charge plotted against t. Preliminary results seem to confirm the experiments on thermoluminescence, and it is hoped to publish a full account of the results at some later date.

CONCLUSIONS

It has been shown that the technique of using a high intensity light flash is particularly effective in removing space charge from a diamond crystal counter when the applied field is switched off. Alternating the applied field with light flashes results in a steady counting rate. By using high field strengths the counting rate remains almost steady until a fairly large space charge field has been built up and hence, if having reached this point a high intensity flash of light is given to the crystal in the absence of the field, the counter is again ready to count at the same rate as before. In this way the counter can be left in an operating condition for a high proportion of the time. Allowance can usually be made for counts missed during the light flash.

In the opinion of the author, this method is more simple and convenient in most cases than the alternating field technique of McKay. However, in some respects, the two techniques are complementary for studying the solid state, e.g., McKay can study only short time changes in the space charge field, whereas the pulsing method can study the long period changes. Both methods are equally satisfactory for studying the behavior under space charge free conditions. The apparatus used by the author was basically simple in design although many improvements may be suggested. In particular, it might be advantageous to control the pulsing circuits electronically; the light could possibly be controlled by a Kerr cell shutter, and in this way the cycle could be performed at much higher rates than when controlled mechanically. This could lead to a more sensitive apparatus for studying the behavior of the space charge.

Experiments of the type described can lead to information concerning the trapping and release of electrons and holes inside the crystal. A simple theory was suggested in order to account for, at least qualitatively, the observed behavior of the space charge although the experimental results show that the actual processes are rather more complex than indicated.

Promising possibilities are suggested by the experiments on the variation of counting rate with temperature. As proposed, increasing the temperature of the crystal to about 600°K should be sufficient to prevent the space charge from forming, and it might thus be possible to obtain a continuously sensitive counter by this means. Possibly, diamond is the only crystal yet studied as a crystal counter which might still function at high temperatures.

Besides the interest in its temperature behavior from the point of view of its possible use as a counter, it has been shown how investigations of the variation of the space charge field with temperature can be made and how the trap depths in the crystal may be estimated. It would be interesting to find how the trap depths obtained in this way compare with those found from the thermoluminescence curves.

Concluding, although the above study of the problems of crystal counters has not resulted in a very practical instrument, it has suggested a new way of approaching the problems of the solid state. Unfortunately, it is rather limited in scope by severe restrictions imposed by the requirements of crystals if they are to be suitable for these types of experiment, i.e., in particular they must remain insulators, preferably over a wide range of temperature. Further, they must respond to individual beta-particles, or they must exhibit a current when bombarded by intense beams of electrons. Also it is desirable for the crystal to be as transparent as possible.

In the next paper, the behavior of the crystal under conditions of freedom from space charge will be described.

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