Behavior of Space Charge Free Diamond Crystal Counters under Beta-Ray Bombardment. II*

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By using the technique described in the preceding paper a study is made of the behavior of a diamond crystal counter under space charge free conditions when subjected to bombardment by beta-rays. The pulse heights caused by the beta-particles are found to be proportional to the energy of the beta-particles over the range 0.66 Mev to 1.0 Mev when a low field strength is used. The trap density in the crystal is found to be of the order of 2×10^{16} per cc and the mean energy required to form an electron hole pair is approximately 7 ev.

THEORY

PAPER I[1] described how the behavior of a diamond crystal counter could be studied under conditions of freedom from space charge by applying an electric field and illumination alternately to the crystal, the light intensity being sufficiently great to remove during the illumination period all the space charge formed in the preceding field pulse. Furthermore, if the rate of formation of space charge due to the beta-rays is not too great, the crystal remains relatively free from space charge throughout each field pulse.

In formulating the behavior of the counter, various assumptions are made; as these have been given in detail by McKay,[2] they will not be repeated here. Let λ_+, λ_- represent the mean free paths of the holes and the secondary electrons in the crystal lattice. Then it can be shown that at low fields when all the freed electrons and holes terminate their paths inside the crystal, the recorded pulse height is

$$q=ne\{(\lambda_++\lambda_-)/d\}, \tag{1}$$

where n is the number of electron hole pairs formed, e is the electronic charge, and d is the distance between the electrodes. The mean free paths are given by the expression,

$$\lambda_\pm=k_\pm FT_\pm, \tag{2}$$

where k_+ and k_- are the mobilities of the holes and electrons, F is the applied field strength, and $T_\pm$ is the time for which the holes and electrons are free.

For high fields or when the beta-ray strikes the crystal near to an electrode, the expression for the mean path has to be modified, since some electrons and holes will have their paths interrupted by reaching the electrodes. It can be shown[3] that the new mean free paths $\bar{\lambda}_+$ and $\bar{\lambda}_-$ are given by,

$$\left.\begin{aligned}\bar{\lambda}_+&=\lambda_+\{1-\exp[-(d-x)/\lambda_+]\}\\ \bar{\lambda}_-&=\lambda_-\{1-\exp(-x/\lambda_-)\}\end{aligned}\right\}, \tag{3}$$

where x is the distance between the anode and the place of formation of the secondary electrons and holes. From Eqs. (1) and (3) the pulse height is now given by

$$q/ne=(\lambda_+/d)\{1-\exp[-(d-x)/\lambda_+]\}+(\lambda_-/d)\{1-\exp(-x/\lambda_-)\}. \tag{4}$$

This assumes that, as was the case, the time constants of the amplifier are large compared with the transit times of the electrons and holes. The expression shows how the recorded charge pulse q varies with the applied field (since $\lambda_\pm$ is proportional to the field) and with the position (x) of the track of the beta-particle. Similar expressions showing these variations have been plotted by Corson and Wilson.[4]

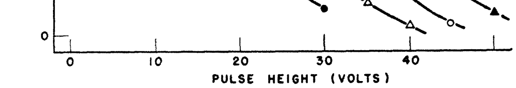

FIG. 1. Curves showing the distribution in pulse heights at various applied fields.

EXPERIMENTAL INVESTIGATION OF THE PROPORTIONALITY OF THE RESPONSE OF THE COUNTER TO β-RAYS

Experimentally the field across the crystal was kept at a known value while a curve was obtained showing the counting rate against the setting of the pulse height discriminator (the bias curve). Differentiation of these curves yielded the counting rate against pulse height curves; these are shown in Fig. 1 for a number of different field strengths. If the counter were behaving

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[1] A. G. Chynoweth, Phys. Rev. 83, 254 (1951).
[2] K. G. McKay, Phys. Rev. 74, 1606 (1948); and 77, 816 (1950).
[3] See N. F. Mott and R. W. Gurney, *Electronic Processes in Ionic Crystals* (Oxford University Press, New York, 1948), p. 122.
[4] D. R. Corson and R. R. Wilson, Rev. Sci. Instr. 19, 207 (1948).

TABLE I. Energy lost by 1-Mev beta-particle in traversing different thicknesses of absorber.

Absorber thickness mg/cm²	Extrapolated max. pulse height volts	Energy lost Mev
0	55.0	0
56.1	54.8	0.004
139.1	53.3	0.031
190.9	50.7	0.078
244.0	48.0	0.127
300.0	39.5	0.282
330.0	35.0	0.364

in a proportional manner, these curves should represent the energy spectrum of the beta-rays striking the crystal. From the foregoing considerations if the field is sufficiently small for all the electrons and holes to be trapped within the crystal, the pulse height is approximately independent of x for $(d-\bar{\lambda}_+)>x>\bar{\lambda}_-$. If $d\gg\lambda_+$ and λ_-, the pulse height is therefore constant for most values of x; and, assuming that the number of electron hole pairs formed is proportional to the energy of the beta-particle, the counter would then behave in an approximately proportional manner.

On the other hand, if the field were sufficiently great for all the electrons and holes to reach the electrodes, the pulse height would then become $q=ne$, and again the counter would behave proportionally. For intermediate fields the pulse height would depend very largely on x and the response would not be proportional to the energy of the β-rays. Summarizing, the differential bias curve should approximate to the energy spectrum of the beta-rays when the applied field is either very low or very high.

At first sight the curves for the lower fields shown in Fig. 1 resemble the usual shape of the beta-ray energy spectrum, the maximum intensity occurring for an energy approximately one-third of the maximum beta-ray energy. However, this is probably due to the accidental canceling of errors as a large number of extraneous effects were present. As very small fields could not be used, since the pulse heights would then be little greater than the noise level of the amplifier, the counter could not be strictly proportional. Also, many of the beta-particles striking the crystal will have already suffered scattering with a consequent loss of energy either in the source or by apparatus surrounding the crystal. Some will be scattered out of the crystal before giving up all their energy, and some energy will be lost by radiation in the lattice field. The actual design of the counter introduces other errors; i.e., some particles will strike the rough edges of the crystal between the electrodes. Also, the higher energy particles will penetrate more deeply into the crystal where the field is a little weaker as a result of distortion, and this will result in too low a pulse height. The net result of these factors will be a preponderance of the lower pulse heights. Finally, there are the statistical fluctuations in the number of beta-particles striking the crystal in a given time and in the number of secondary electrons that they create. Counting errors are present; also slight fluctuations in the noise level of the amplifier will occur, thereby causing a similar fluctuation in otherwise equal pulse heights. The latter effect will be relatively more serious for the smaller pulses such as at small field strengths.

As very high field strengths could not be used because of spurious pulses arising in the counter from charge leaks, the proportionality of the counter was not tested in this region.

In consequence of the foregoing considerations a new approach was made in order to test the proportionality of the counter at low fields. It is obvious that irrespective of the shape of the beta-ray spectrum, the largest pulse (at low fields) should represent those particles of highest energy which have given up the whole of their energy to the crystal lattice. Hence, by interposing metal absorbers of various thicknesses in the beta-ray beam, the maximum incident beta-ray energies can be varied and the effect on the maximum pulse size studied.

With the beta-rays from RaE (maximum energy approximately 1 Mev) bias curves were obtained with aluminum absorbers of thicknesses ranging up to about 300 mg/cm². For the thicker absorbers a stronger source was used; but even so, curves could not be obtained for thicknesses greater than 300 mg/cm², as the signal-to-noise ratio became too low. The bias curves were differentiated and extrapolated in order to find the maximum pulse height corresponding to each absorber. These extrapolated heights are given in Table I.

With no absorber present the pulse height was 55 volts, and taking this to represent beta-particles of 1 Mev energy the energy lost by these beta-particles in traversing the various absorbers was determined by assuming that the response of the counter was proportional to the energy of the beta-particles. This energy

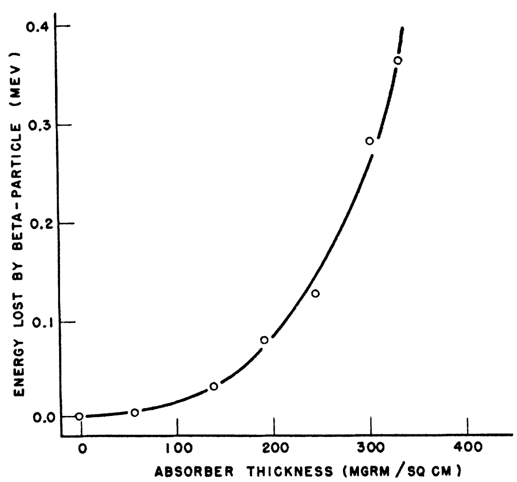

FIG. 2. Curve showing the energy lost by a 1-Mev beta-particle in traversing various thicknesses of aluminum as found experimentally.

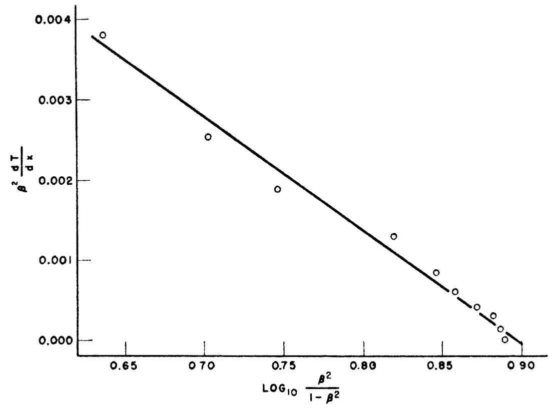

FIG. 3. Plot of $\beta^2 dT/dx$ against $\log[\beta^2/(1-\beta^2)]$ where β is the ratio of the velocity of the beta-particle to that of light and dT is the energy lost by the beta-particle in traversing a thickness dx of the absorber.

loss is given in the final column of Table I. It is plotted in Fig. 2 against the absorber thickness.

Bohr has shown that the rate of loss of energy by a charged particle in traversing a thickness x of material is given by

$$\frac{dT}{dx}=\frac{4\pi e^4 Z^2}{mv^2} n\sum_i f_i \log_e\left\{\frac{mv^2}{\hbar\omega_i[1-(v^2/c^2)]}\right\},$$

where m is the rest mass of the electron, v is the velocity of the beta-particle, ω_i is the vibration frequency of the ith atomic electron, n is the number of atoms/cc, Z is the number of electron charges in the bombarding particle, and f_i is the oscillator strength corresponding to the ith electron. Hence, if $\beta=v/c$, a graph of $\beta^2 dT/dx$ against $\log[\beta^2/(1-\beta^2)]$ should be a straight line.

From the curve of Fig. 2 the values of dT/dx corresponding to various values of x were obtained and also the corresponding values of T, the energy of the beta-particle. Knowing T, β was determined and hence $\log[\beta^2/(1-\beta^2)]$ and $\beta^2 dT/dx$. These are shown plotted

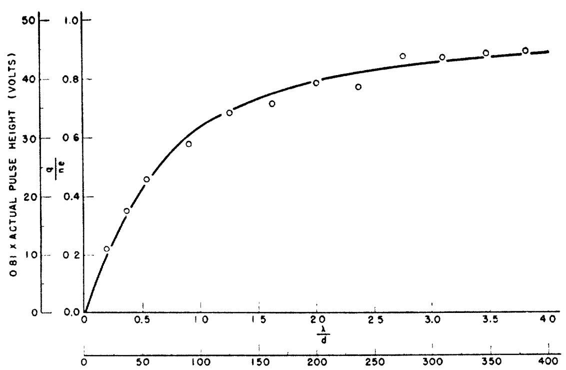

FIG. 4. The full curve shows the theoretical variation of pulse height with field strength (λ/d is proportional to the field). The plotted points are those obtained experimentally; they are fitted to the theoretical curve by means of the scaling factors given on the axes.

in Fig. 3, and it is seen that the relation is reasonably linear over the range of values of dT/dx that it was possible to obtain. Besides the errors involved in the experiment already referred to, it is not accurate to assume that the highest energy beta-particles striking the crystal are 1-Mev particles. The upper limit to the beta-ray spectrum is 1.05 Mev, and it is probable that the energy lost by the beta-particle in passing through the thin cover over the source was rather less than 0.05 Mev. If this were the case, it would lead to too low an experimental value for β and this would cause a deviation in the direction observed for small absorber thicknesses. Further errors are introduced, since, strictly, the equation applies only to absorbers formed of heavy atoms where the orbital electrons can be treated statistically as a gas. Finally, it has been tacitly assumed throughout that the energy required to liberate an electron from the crystal lattice is independent of the energy of the beta-particles. However, it is felt that the plot shown in Fig. 3 is strong evidence for the proportional behavior of the counter for β-particles of energies between 0.66 and 1 Mev.

VARIATION OF PULSE HEIGHT WITH FIELD

By extrapolating the bias curves obtained for various applied fields, the variation of the maximum pulse height with field could be obtained. The theoretical variation of the pulse height can be obtained as follows: Eq. (4) shows the variation of q with x and for the maximum pulse height,

$$d/dx(q/ne)=0.$$

This gives the condition for maximum pulse height, i.e., when

$$x=d\lambda_-/(\lambda_+ +\lambda_-),$$

and this is independent of the field strength. Substituting this value for x into Eq. (4) gives

$$q_{\max}/ne=(\lambda_- +\lambda_+/d)\{1-\exp[-d/(\lambda_+ +\lambda_-)]\},$$

or writing $\lambda=\lambda_- +\lambda_+$,

$$q_{\max}/ne=\lambda/d[1-\exp(-d/\lambda)].$$

This describes the variation of the maximum pulse height with the applied field.

In Fig. 4 the theoretical curve of q/ne against arbitrary values of λ/d is plotted. Also shown are the points experimentally obtained, these having been fitted to the theoretical curve by using the scaling factors that are given on the axes. It is seen that the agreement between the theory and the experiment is quite satisfactory.

As was pointed out by McKay,[2] curves of this type yield useful information concerning the crystal; i.e., from the scaling factors the density of trapping states in the crystal can be estimated together with the mean energy required for a β-particle to release a lattice electron.

DENSITY OF TRAPPING STATES

From Fig. 4 it is seen that the factor $(\lambda_- + \lambda_+)/d$ is unity when the applied voltage is 100/0.73; and, hence, knowing the electrode separation to be 0.15 cm, we obtain

$$k_+T_+ + k_-T_- = 16.4\times 10^{-5}\ \mathrm{cm^2/volt}.$$

Several authors have reported the results of experiments to determine the mobilities of electrons and holes in diamond. McKay in his second paper obtained values of 8.3×10^{-6} and 4.6×10^{-6} cm^2/volt for k_+T_+ and k_-T_- respectively.' More recently, Pearlstein and Sutton[5] using a method similar to that of McKay found these values to be 3.5×10^{-5} cm^2/volt and 3.8×10^{-5} cm^2/volt, respectively. Finally Freeman and van der Velden[6] have described experiments somewhat similar to those of the author's (except that α-rays were used) and a result of 5.3×10^{-5} cm^2/volt was obtained. It appears, therefore, that there is quite considerable disagreement amongst authors concerning the value of $k_\pm T_\pm$ and all the experimental results lead to values for the electron mobility considerably different to that deduced theoretically by Seitz,[7] i.e., 156 cm/sec/volt/cm. It seems most likely that the value of $k_\pm T_\pm$ can vary appreciably for different diamond specimens. This is not unexpected, as many of the physical properties of diamonds show marked variation in magnitude from specimen to specimen. Taking the most recent values for $k_\pm T_\pm$, i.e., those of Pearlstein and Sutton, we can write,

$$k_+T_+ \sim k_-T_-,$$

and hence,

$$k_+T_+ = k_-T_- \sim 8.2\times 10^{-5}\ \mathrm{cm^2/volt}.$$

Thus there is fair agreement between the author, Freeman and van der Velden, and Pearlstein and Sutton as to the order of magnitude of kT.

Pearlstein and Sutton were able to measure $T_\pm$ independently and hence could obtain results for $k_\pm$. They were $k_+ = 4800\pm 20$ percent and $k_- = 3900\pm 15$ percent. (For another specimen preliminary experiments gave k_- as 2760 with a rather large expected error.) Klick and Maurer[8] by making Hall effect measurements in a single diamond specimen found the electron mobility to be 900 cm^2/sec/volt though this result is open to some question, as they neglected the movement of the positive holes. Using the more recent values for the mobilities as found by Pearlstein and Sutton, we obtain

$$T_\pm \sim 2\times 10^{-8}\ \mathrm{sec}.$$

Now the trap density N is given by

$$N = 1/T\sigma u,$$

where σ is the capture cross section of the traps and u is the thermal velocity of the free electrons (approximately 1.2×10^7 cm/sec). It has to be assumed that the velocity imparted to the electron by the field is small compared with its thermal velocity; this is true for the lower field strengths used.

As the nature of the trapping mechanism in diamond is not known, we can obtain only an approximate value for σ by comparing the trapping centers with the luminescent centers of ZnS phosphors and similar materials. The value of σ in these studies[3,7] is usually taken to be about 10^{-15} cm^2, and hence we obtain for the density of trapping centers

$$N \sim 2\times 10^{16}/\mathrm{cc}.$$

This figure seems not unreasonable, for it is of the same order as the trap density found in many luminescent and photoconducting materials.[3]

ENERGY PER ELECTRON-HOLE PAIR

From Fig. 4 it is seen that for infinite field strength the output pulse would be about 62 volts, and this corresponds to the total collection of both secondary electrons and positive holes. The gain of the amplifier was measured by putting voltage pulses of known magnitude on to the first grid. The capacity of the input circuit was kept to a minimum by careful construction and it was estimated to be $8\pm 2\ \mu\mu$f. Again assuming the largest pulses to be caused by beta-particles of 1 Mev energy, the mean energy required to form an electron-hole pair was calculated to be about 7 ev. This value is substantially lower than the values previous authors have given. Ahearn's[9] results give the value of 10 ev for the energy required to free an electron by alpha-rays; and McKay estimates that after allowing for the energy absorbed from the low energy electron beams by the metal electrode, his experiments would yield the same value (though with a rather large possible error). The author has found no value in the literature for the liberation energy in diamond for beta-rays of the order of 1 Mev energy. However, these would penetrate through the surface layers of the crystal without any great loss and give up nearly all of their energy to the interior of the crystal. Hence there is no need to make the necessarily highly approximate corrections for energy loss in the electrode. That the effect of the surface layer is considerable for alpha-rays has been shown by van Heerden[10] with silver chloride crystals. Also, Ahearn[11] has investigated the variation in counting efficiency for alpha-particles over various parts of the surface of a diamond crystal finding the best response near to a particularly large surface flaw. Thus it is likely that particles of low penetrating power

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[11] A. J. Ahearn, Rochester Report on High Speed Counters (July, 1948).

will not yield very reliable values for the liberation energy, and it is probable that the figure found by the author for 1-Mev beta-rays is somewhat nearer the true value. Finally, as was pointed out by Freeman and van der Velden,[6] pulse counting methods are more likely than current measuring methods to yield accurate results for measurements of kT and the energy per electron-hole pair. Current methods give a value for kT which is the average over all the electrons in the current. If, owing to the presence of a mechanical flaw or some other cause, some electrons are unable to contribute to the current to the extent that they would be able to if in a normal lattice, the measured current will be less than the value it would have in a normal lattice. This in turn would lead to a higher value for the energy per electron-hole pair. In pulse counting methods it is not necessary to assume that *all* the electrons and holes produced by *all* the beta-particles travel in a normal lattice. It is necessary only to assume that all the electrons and holes produced by *some* beta-particles travel in the normal lattice, and this is obviously a much more reasonable assumption. Summarizing, it seems fairly safe to assume that the maximum observed pulse heights do represent the motion of the electrons and holes in the normal diamond lattice.

CONCLUSIONS

It is hoped to extend the foregoing types of investigation to a number of diamond crystals, and in this way it may be possible to correlate the possible variation in trap density or energy per electron-hole pair with some other physical property. In particular, it has sometimes been suggested that the response of the crystal depends on its structure, i.e., whether it is a laminated, mosaic, or perfect crystal lattice. The degree of imperfection of the structure of the crystal can be determined by means of the divergent beam x-ray technique developed by Lonsdale[12] and such studies may yield information as to whether trapping sites mainly exist at the boundaries of mosaic blocks or laminations; i.e., if the traps do occur at the boundaries, then the trap density should be greater for those diamonds which show the better divergent beam photographs.

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Potential and Gradient Distributions in Parallel Plane Diodes at Currents below Space-Charge-Limited Values

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The equations for electron motion in a plane diode under the influence of space charge are integrated. The solution is expressed in terms of four dimensionless numbers, normalized so that their range is from zero to unity. The gradient at the cathode is expressed as a function of the current density as it varies from zero to space-charge-limited values. Space distributions of potential and gradient are given in terms of the location of the plane of interest between the cathode and the anode for a number of specific current densities.

Evidence is shown of the existence of a plane for which the gradient is essentially independent of the current density.

INTRODUCTION

EXPRESSIONS for the complete description of the potential and gradient distributions in parallel plane diodes under conditions of emission-limited currents are given in terms of four dimensionless variables.[1] Others[2–6] have given equations for the same phenomena, but those of which the author is aware all make use of parameters whose physical significance is very obscure. The variables used in this derivation represent current density, voltage, distance, and cathode gradient; normalization is such that these variables range between zero and unity as the current density varies from zero to space-charge-limited values.

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