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ELECTRONIC PROPERTIES OF PARTICLE-COUNTING DIAMONDS
II. PARTICLE-COUNTING PROPERTIES

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CONTENTS

A survey of the counting characteristics of thirty particle-counting diamonds has been made. It was found that the diamonds can be classified on the basis of their counting behaviour in the dark and during subsequent illumination. The dark characteristics and the counting response to illumination are explained in terms of the trapping and recombination of charge carriers via three levels in the forbidden energy gap. It is shown formally that the three-level model predicts the empirical classification of the diamonds on the basis of varying concentrations of the defects that make the diamonds be of type Ia. The asymmetry of the counting property is due to inhomogeneity of the defects and the counting property can be put into correspondence with the purity of a diamond, the purest diamonds being the best counters.

1. General introduction

The stimulus for the investigation of the counting properties of diamonds has come from their possible use as crystal counters in nuclear physics research. It is found that the properties of counting diamonds are greatly variable, and in general they do not exhibit the desired energy proportionality of good crystal counters (Champion & Wright 1959). However, the counting property serves as a means whereby electronic transitions between energy bands and isolated levels can be studied.

In these experiments a diamond is held between two electrodes and an electric field is applied across it. The ionizing radiation which penetrates the crystal generates small conduction pulses. These are amplified and studied as functions of any of several variables such as the time, the electric field strength and the illumination of the crystal.

Counting diamonds are rare, and considerable effort has been devoted to correlating the counting property with other physical properties of diamond (Ahearn 1948; Friedman, Birks & Gauvin 1948; Lonsdale 1948; Hofstadter 1948; Freeman & van der Velden 1951*a*; Champion 1952, 1953, 1956; Frerichs 1950; Taylor 1956; Champion & Kennedy 1965). The relation between the counting property and the type and purity of a diamond is not clear.

The present paper describes an investigation into the β-particle counting properties of thirty 'good' counting diamonds in which most of the results previously obtained by other workers were reproduced. An empirical classification of the counting diamonds has resulted. It has also become evident that the time dependence of the dark counting rate is correlated with the subsequent response of the counting rate to illumination. Most diamonds exhibit an asymmetry in their counting efficiency with respect to reversal of the applied field.

In a search for the physical reasons underlying the empirical classification of the counting diamonds, the optical absorption and photoconduction spectra of our counting diamonds and of numbers of other diamonds of all the known types were studied. This work was reported in part I (preceding paper). The photoconduction spectra of the counting diamonds were interpreted in part I in terms of electronic transitions between the energy bands and three isolated levels at 0·85, 1·45 and 2·2 eV from the valence band.

This model was shown to be consistent with all the electro-optical phenomena in type Ia diamonds of which we are aware. It will be further analysed in the present paper. The further analysis strongly suggests that the physical basis for the classification of the counting diamonds is provided by variations in the relative concentrations of the defects that make these diamonds be of type Ia. With this addition to the existing theory, it is found possible to understand the relationship between the time dependence of the dark count rate of a diamond and its counting response to illumination. Also, it is possible to put the counting efficiency of a diamond into correspondence with its purity—the purest diamonds being the best counters. The asymmetry of the counting property in diamonds can then be explained in terms of an inhomogeneous distribution of the defects.

2. Introduction

Diamonds were divided into two types by Robertson, Fox & Martin (1934) on the basis of differences in their electrical and optical properties. Type II diamonds were transparent in the ultraviolet between 3000 and 2250 Å and more strongly photoconducting than type I. We will introduce a further analysis of diamonds into various *classes* and *groups*. This analysis is based on their counting properties alone and is not to be confused with the division of diamonds into *types* by Robertson *et al.*

The behaviour of counting diamonds has now been studied by a number of authors (Wooldridge, Ahearn & Burton 1947; McKay 1948, 1950; Chynoweth 1949, 1951*a*, *b*; Freeman & van der Velden 1950, 1951*b*, 1952*a*, *b*; Willardson & Danielson 1950*a*, *b*, 1952; Stratton & Champion 1952; Trott 1953; Champion & Dale 1956; Urlau, Logie & Nabarro 1961 and others). The investigations were mostly confined to a few diamonds.

It is found that the counting efficiency of all diamonds varies with time. In some cases it decreases, which indicates that the counting is being limited by the development of space charge. In other cases it increases in time to a steady value, and in yet other cases it first passes through a minimum before levelling off to a steady value.

The response of the counting rate to illumination has also shown wide variations. The majority of counting diamonds have exhibited a positive counting response to red light (i.e. red light increases the counting rate) and a negative response to green light and light

of higher energy. In a few cases, negative responses to both red and green light have been observed. A response may be analysed into transient and permanent parts. The transients may be either positive or negative. All possible conbinations of transients and permanent responses have been observed.

The situation is confused because the investigations have differed in their choice of the variables listed at the beginning of § 4. Also, it has to be kept in mind that every diamond is an individual. It is not clear whether the diversity of results that has been reported is due to variation in the experimental conditions or to differences between the various diamonds.

3. Apparatus used in the counting experiments

A block diagram of this apparatus is shown in figure 1. The scaler, recorder and pulse height analyser are standard equipment. The preamplifier was built according to the circuit of Cottini, Gatti, Gianelli & Rozzi (1956).

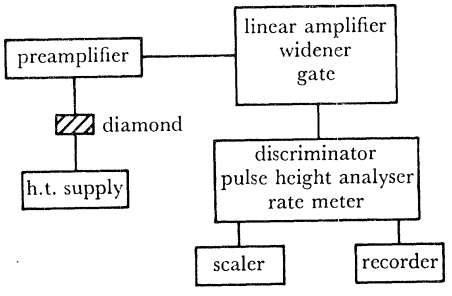


Figure 1. A block diagram of the apparatus used in the counting experiments.

Use was also made of a 512 channel pulse height analyser by Nuclear Data, Chicago.

The preamplifier was linear over a range of input pulse heights from 0 to 16 mV with a gain of about 34. The average output pulse height with the diamond as detector was about 5 mV.

The crystal holder was simply an arrangement whereby the diamond could be clamped between two electrodes. It was mounted on the preamplifier chassis so as to keep the signal-carrying lead as short as possible. The crystal holder was surrounded by a light-tight box provided with a window, shutter and light pipe so that the diamonds could be kept either in darkness or illuminated with the desired light.

The counting diamonds were octahedral crystals of the order of 1 mm^3.

The recorded counting rate comprised all pulses above the noise level, which was discriminated against by the threshold bias of the discriminator. Pulse spectra obtained with the single-channel pulse height analyser will be called 'bias spectra'.

For illumination of the diamonds, monochromatic light was filtered from a white light beam by means of interference filters. The intensity of the light incident on a diamond in the crystal holder was measured with a thermopile, and intensity of illumination ($3{\cdot}3 \times 10^{12}$ photons $cm^{-2}s^{-1}$) was kept constant, independent of the wavelength.

4. The experimental routine for the counting experiments

The counting properties of diamonds may be studied under a variety of conditions. We list some of the experimental variables and possible experiments.

1. The nature of the ionizing radiation may be varied.

2. The counting efficiency may be studied as a function of the time and of the field strength.

3. The direction of bombardment may be varied—it may be either parallel or perpendicular to the applied field.

4. The diamond may be kept in darkness or illuminated. The wavelength and intensity of the illumination may be varied.

5. The decay of the continued counting after the applied field has been switched off may be studied.

A complete investigation would require that each of the experiments derived through permutation of (1) to (5) be carried out for every stone. The experiments are very time-consuming and the results are in general not reproducible unless the diamonds are returned to some sort of standard state.

The experimental conditions and the routine followed in the present work were arrived at after an extensive preliminary investigation of the counting properties of a number of diamonds and a comparison of the results obtained with those previously reported by other workers.

It was decided:

1. To bombard with β-particles from a $^{90}Sr^{90}Y$ β source with reproducible source geometry.

2. To bombard the diamonds uniformly and only perpendicular to the applied field.

3. To use a single low field strength for all the diamonds, which would be high enough to ensure that all of the so-called good counting diamonds would yield appreciable counting rates, but low enough to ensure good sensitivity to red light. The preliminary work indicated that diamonds which exhibit a positive counting response to red light are most sensitive when the applied field is low. A field strength of 5000 V/cm was suitable.

The standard state referred to above was achieved by leaving the crystals for a sufficiently long time in darkness, without an applied field, and under continuous bombardment with β-particles. This operation will be called 'blacking-out' the crystal.

Whether or not a diamond was in the standard state at the commencement of observations was inferred from the reproducibility of the initial and steady values of its counting rate in the dark. After a black-out time of 6 h these quantities were reproducible to within 2 and 5 % respectively in all the counting diamonds.

After blacking-out, the counting response of a diamond to illumination, once it is in its steady counting state, is fully reproducible. When the illumination was switched off, the previous steady dark counting rate was in most cases recoverable and the counting response to illumination reproducible.

The experimental routine adopted for the present work was as follows:

1. Black-out the crystal for 6 h.

2. Switch on the field and record the characteristic of dark counting rate against time; wait until the steady counting state has been reached.

3. Illuminate and record the counting response to illumination; wait until the steady counting state during illumination has been attained.

4. Switch off the light and record the recovery; wait until the previous steady dark counting rate has been recovered.

5. Test for reproducibility.

6. Repeat the cycle with illumination of the next colour.

Illumination always began with light of the lowest energy.

In addition to the above routine, measurements were made on some diamonds of the decay of the continued counting rate after the applied field was switched off and of the count rate/voltage characteristics.

5. Results

5·1. *Dark counting*

The count rate/voltage characteristics were as found by Willardson & Danielson (1950*a*, *b*; 1952), Kojima & Kono (1952), Urlau *et al.* (1961) and others, and shown in figure 2. The counting rate tends to a saturation value at high applied fields.

The dark count rate/time characteristics were of three classes, as shown in figure 3. After the occurrence of an initial maximum (not shown in figure 3) which is only developed in some diamonds, the counting rate either:

(*a*) decreased monotonically to a steady value, or

(*b*) passed through a minimum before levelling off to a steady value.

The two classes of characteristic are labelled respectively class A and class B. Class B is further subdivided. If the initial counting rate is higher than the final steady value, the diamond is called class B_1. If is is lower than the final steady value, the diamond is called class B_2.

In the present work:

9 diamonds were of class A;
17 diamonds were of class B_1;
4 diamonds were of class B_2.

The time required to reach the steady counting rate ranged from 2 to 27 h.

The class A characteristic is similar to that found by Freeman & van der Velden (1950, 1951*b*, 1952*a*, *b*), Chynoweth (1951 *a*, *b*), Champion & Dale (1956), Urlau *et al.* (1961) and others.

The class B_1 characteristic has been reported by Stratton & Champion (1952) and Trott (1953) and the class B_2 characteristic was previously seen by Willardson & Danielson (1950*a*, *b*; 1952).

The depolarization counting of some of the diamonds of all classes was also studied. The procedure was to black out the crystal, switch on the field to obtain the dark characteristic,

and then switch off the field to obtain the depolarization characteristic. The polarity of the pulses was then reversed, since the diamond now counted under its own space charge field.

It was found:

(i) in agreement with Urlau (1960) and others, that the depolarization characteristic was compound exponential, i.e. of the form

$$\text{counting rate} = a_1 \mathrm{e}^{-\lambda_1 t} + a_2 \mathrm{e}^{-\lambda_2 t} + \ldots, \tag{1}$$

where the a_i and λ_i are positive constants and t the time.

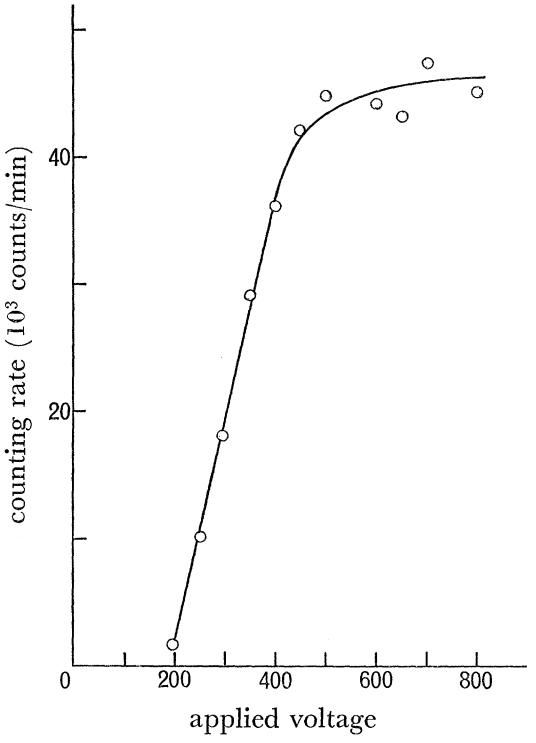


FIGURE 2. The counting rate/voltage characteristic of diamond C2.

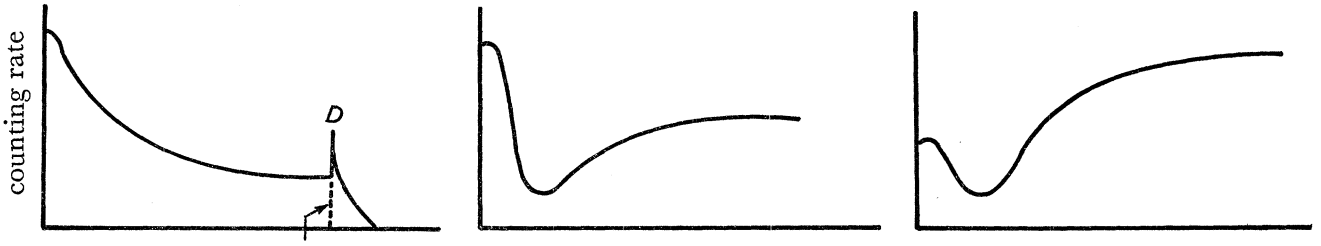
(ii) that sometimes a peak could be seen when the field is switched off. This is shown in figure 3(*a*), where it is labelled D.

This effect was previously seen by Urlau (1960) on only one diamond, and interpreted by him as being due to an increase in the lifetime of the charge carriers. In the present work, the occurrence of this peak was found to be voltage dependent, and it could be seen in the count rate/time characteristic obtained from every diamond investigated. The magnitude of the peak above the previous steady dark count rate was largest when the previously applied field was small, decreasing steadily as the field increased.

The pulses at the output of the preamplifier were also observed on an oscilloscope screen. With the applied field on, they were negative-going pulses—so arranged to meet the input requirements of the main amplifier. When the applied field was switched off, the vast majority were positive, but a few negative-going pulses were still to be seen.

Different steady counting rates were obtained when the crystal orientation in the applied field was altered or the applied field simply reversed. All of the diamonds examined here

exhibited this property. The phenomenon was previously reported by Stratton & Champion (1952) and Taylor (1956). The effect was not studied in detail. The orientation of the diamond could also govern the class of dark characteristic associated with the crystal.



(*a*) class A (*b*) class B_1 (*c*) class B_2

FIGURE 3. The counting rate/time characteristics.

5·2. *Counting during illumination*

The routine outlined in § 4 was followed. It was found that the effect of illumination by light of any energy between 1·6 and 2·0 eV was qualitatively the same. This will be called the response to red light. If the energy exceeded about 2·2 eV, the nature of the response changed to what will be called the green-light response. The transition occurred in the neighbourhood of 2·2 eV for all the diamonds. The diamonds were divided into two *groups* according to whether the red-light response was positive or negative. A response is called positive if the final count rate under illumination is greater than that in the dark and negative if it is less.

Group A consists of all those diamonds whose red-light response is positive. It includes a number of crystals which are anomalous and which will be discussed separately in § 5·2·3.

Group B consists of all those diamonds whose count rate response to red light is negative.

5·2·1. *The diamonds of group A*

Under conditions where the applied field is 5 kV/cm and the bombardment is with β-particles perpendicular to the applied field, these diamonds:

1. All respond positively to red light.
2. Have dark count rate/time characteristics of either class A or class B_1.
3. Have a counting response to green light which may be either positive or negative. For the negative-response crystals the critical energy, although not very well defined, is in the neighbourhood of 2·2 eV. The positive response crystals have interesting properties, and will be discussed separately.

As an example of the 'ordinary' diamonds of group A we will consider the results obtained from diamond G 2. The complete plot of its counting rate in time is shown in figure 4. The figure also demonstrates the routine according to which the observations were obtained.

The tests of the reproducibility of the counting response to 587 nm illumination are shown in figure 5.

Re-illumination after a recovery time of only three hours resulted in the response of figure 5 (*a*). If the dark time was 12 h or longer, then the response was fully reproducible as shown in figure 5 (*b*).

Typical results for the group A diamonds are summarized in figure 6. The transients associated with the response and recovery are also shown. In many cases they can be interpreted.

That the effect of the illumination is to change the overall pulse height is shown in figures 7 and 8. Figure 7 was obtained with a multichannel pulse height analyser.

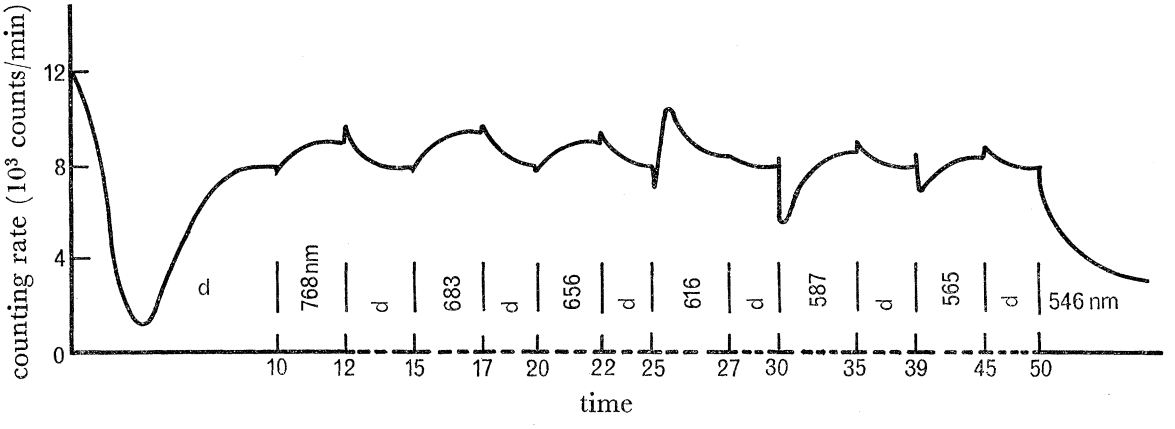


FIGURE 4. Variation of the counting rate when diamond G2 is illuminated with monochromatic light.

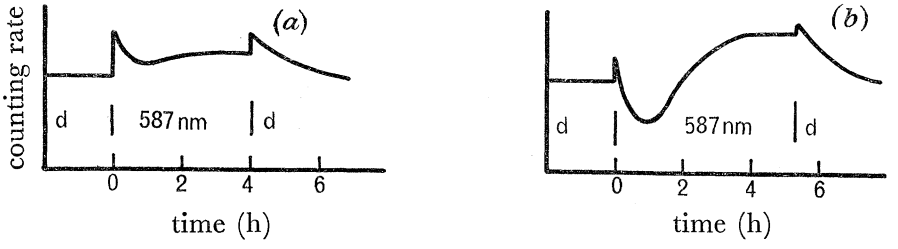


FIGURE 5*a*. The response of G2 to 587 nm after a recovery time of 3 h.

FIGURE 5*b*. The response of G2 to 587 nm after a recovery time of 12 h.

Figure 8 shows the bias spectra of the pulses obtained from diamond C2 with only 200 V applied across the crystal. It is the plot of the number of pulses between T and $T+\Delta T$ where T is the value of the threshold bias (in divisions) and ΔT the window width. It demonstrates the large effect that red light can have on the counting when the applied field is low. Green light eliminates the counting under the same conditions.

5·2·2. *The diamonds of group B*

These are the crystals whose count rate response to red light is negative. The special properties of this group are illustrated qualitatively in figure 9. The characteristic feature is that the largest negative response is obtained with light of the lowest energy, the magnitude of the response steadily decreasing as the energy of the light is increased until a critical energy is reached when the response is again large and negative. The critical energy was 2·2 eV for all the diamonds of this group. Typical responses to monochromatic light for this group are collected in figure 10. The magnitude of the responses relative to the steady dark count rates are given whenever possible.

crystal	class	fluo-rescence	response of group A to monochromatic illumination						transition energy-eV
			1·6 eV	1·8 eV	1·9 eV	2·1 eV	2·2 eV	2·4 eV	
C2	B_1	no							2·2
C3	A	no							2·1
C8	A	no	16%	50		−3			2·1
G2	B_1	no	20	51	45	28	75		2·1–2·2
G5	B_1	yes							2·1–2·2
G7	A	yes							
G8	B_1	yes	46	110	110	63	5	−50	2·2–2·4
G10	A	yes							
G11	A	yes	90	110	100	100	50	−23	2·1–2·4
G12	A	yes							
G15	B_1	no	8			16	−5	−80	2·2
G21	A	no							2·2

FIGURE 6. The counting response of group A diamonds to monochromatic illumination.

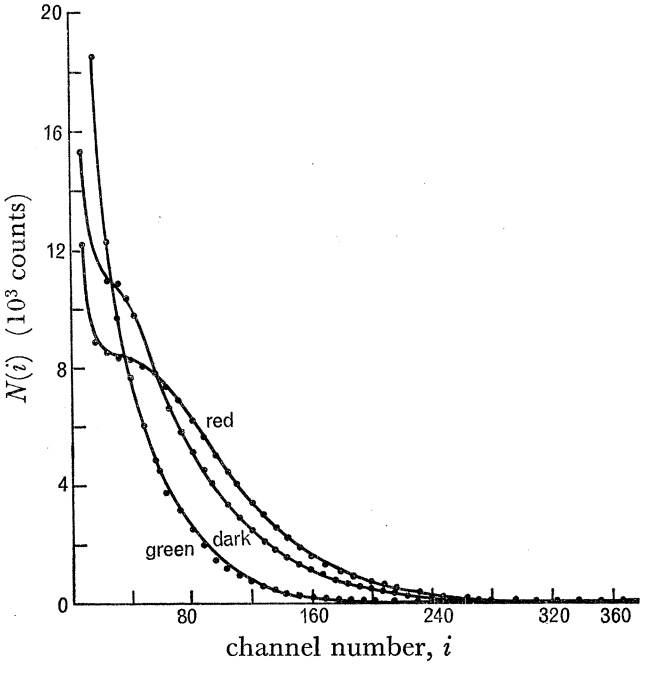


FIGURE 7. The spectra of 10^6 pulses obtained from diamond G21 as measured with a multichannel pulse height analyser. The photon flux was the same during illumination with red (683 nm) and green (523 nm) light.

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5·2·3. *The anomalous diamonds of group A*

These diamonds have the following properties:

1. Their dark count rate characteristics are class A with very high initial counting rates but with a steady counting rate of only a few hundred counts per minute. The steady count rate is reached within a few hours of dark counting.

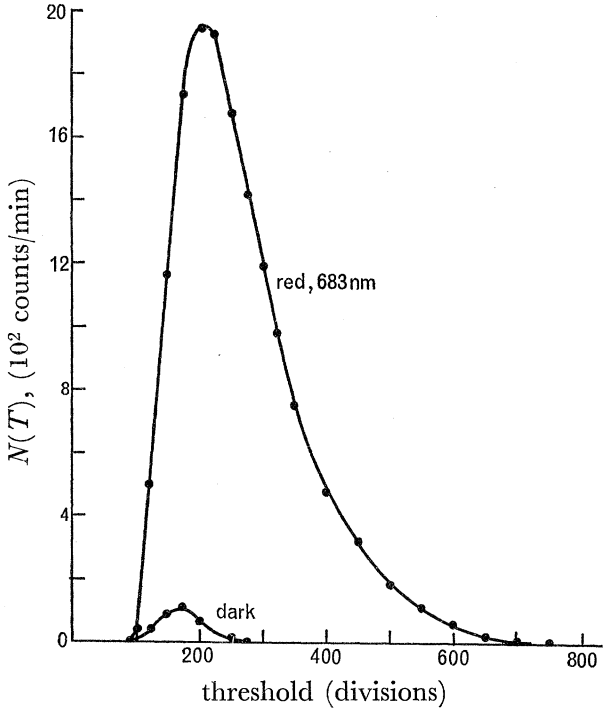

FIGURE 8. The bias spectra of the pulses from diamond C2 when 200 V was applied to the crystal. The window width was 10 divisions.

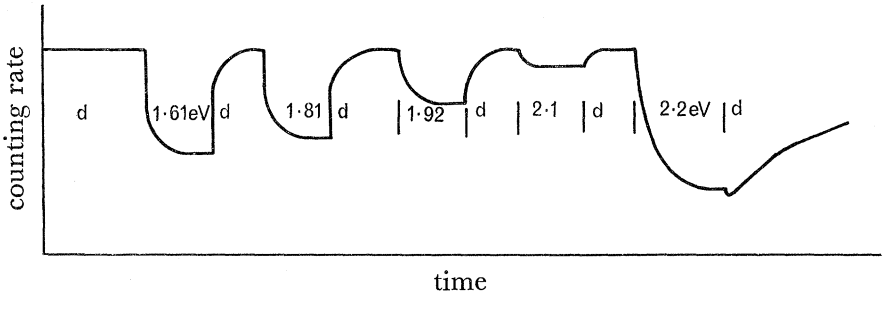

FIGURE 9. A qualitative representation of the counting response of group B diamonds to monochromatic light.

2. Their response to red light is large, positive, transient and fully reproducible after about 10 h dark counting. As the energy of the light is increased so the magnitude of the transient is decreased and the magnitude of the response increased.

3. The response to green light of all energies greater than 2·0 eV is still positive. In this respect these diamonds are different from all the others. For all the other diamonds an

crystal	class	fluo-rescence	response of group B to monochromatic illumination						critical energy-eV
			1·6 eV	1·8 eV	1·9 eV	2·1 eV	2·2 eV	2·4 eV	
C7	B_2	no	−40%	−35	−26	−19	−51	−71	2·2
G1	B_2	no	−35	−25	−15	−10	−40		2·2
G3	B_2	no	−40	−33		−10	−45		2·2
G4	B_2	no	−8	−8	−8	0		−32	2·2
G6	B_2	yes	−5	−5	−3	−3	−40	−70	2·2
G9	B_1	yes	−25	−22	−15	−5	−53	−98	2·2

FIGURE 10. The counting responses of group B diamonds to monochromatic illumination.

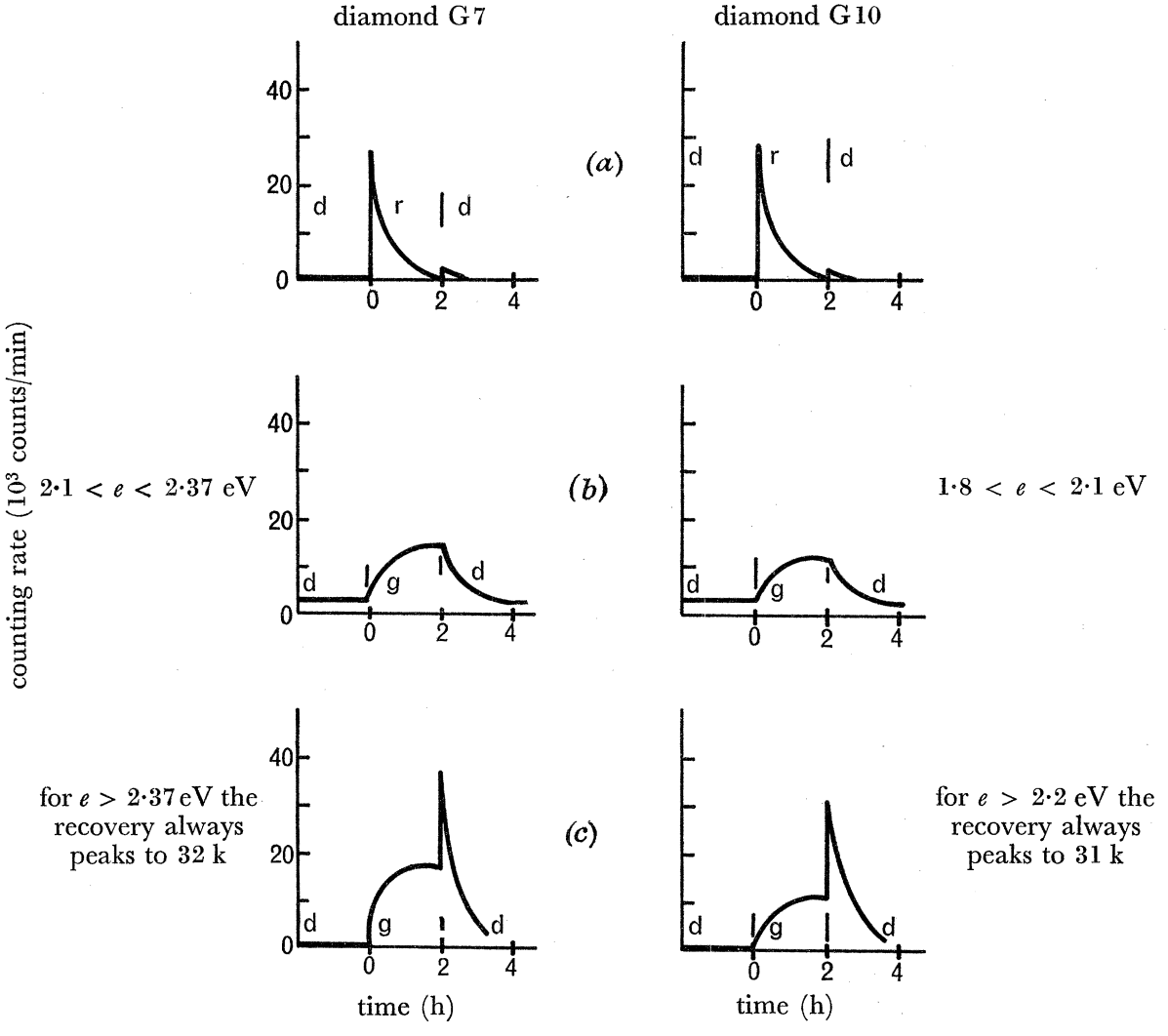

FIGURE 11. The counting response of diamonds G7 and G10 to (*a*) red light, (*b*) green light, and (*c*) green light.

energy in the range 2·1 to 2·4 eV can always be found which will have a large depressant effect on the counting.

Typical of these diamonds are G7 and G10, whose very similar responses are shown in figure 11.

6. Theory and discussion of the counting properties of diamond

We first summarize the properties of the groups. Group A is divided into two subgroups called 'ordinary' and 'anomalous'. The majority are ordinary.

Group A ordinary

The counting response to red light is positive.
The counting response to green light is negative.
The dark characteristic is either class A or class B_1.

Group A anomalous

The counting response to red light is positive.
The counting response to green light is positive.
The dark characteristic is class A.

Group B

The counting response to red light is negative.
The counting response to green light is negative.
The dark characteristic is class B_1 or class B_2.

We notice, and this is purely empirical, that the diamonds of class A are always of group A and the diamonds of class B_2 are always of group B. This qualitative correlation between the classes and the groups is confirmed by statistical analysis on the basis that the thirty diamonds examined here constitute a random sample of counting diamonds. Let P_A be the proportion of all diamonds of class A that are of group A, and P_{B_2} be the proportion of all diamonds of class B_2 that are of group A. The probability of finding nine out of nine diamonds of class A with group A behaviour is less than 0·025 if $P_A < 0{\cdot}664$. The probability of finding four out of four diamonds of class B_2 with group B behaviour is less than 0·025 if $P_{B_2} > 0{\cdot}602$. This suggests strongly that $0{\cdot}664 < P_A < 1$ and $0 < P_{B_2} < 0{\cdot}602$, and it is significant that the range of P_A does not overlap that of P_{B_2}. It may be judged with a high degree of confidence that $P_A > P_{B_2}$, that is, that the proportion of diamonds of class A with group A behaviour is greater than the proportion of diamonds in class B_2 with group A behaviour.

To obtain further guidance in this complicated situation we summarize in table 1 the related observations of other workers.

In column 2 is reported where possible:

(1) The nature of the ionizing radiation.

(2) The direction of bombardment, whether parallel or perpendicular to the applied field.

In column 3 is reported:

(1) The class of dark count rate/time characteristic.

(2) The effect of red light.

(3) The effect of any other light.

These results are all consistent with the empirical relation between the classes and the groups.

TABLE 1

author	experimental details	observations
Chynoweth (1949, 1951 *a*)	(1) α and β particles (2) perpendicular to the applied field	(1) class A (2) positive to red light
Freeman & van der Velden (1950)	(1) α particles (2) bombardment through cathode	(1) class A (2) positive to red light
Willardson & Danielson (1950 *b*, 1952)	(1) γ rays (2) perpendicular to the field	(1) class A and class B (2) positive and negative to red light
These authors report on the 'activation' of a diamond. In the 'activated state' the response to red light is negative.		
Kojima & Kono (1952)	(1) α particles (2*a*) uncollimated beam perpendicular to the applied field (2*b*) collimated α particle beam perpendicular to the applied field saturating field strengths employed	(1) class A (2) negative to red light (3) negative to green and white light (2) positive response to red light (3) negative to green and white light
Stratton & Champion (1952)	(1) β particles (2) bombardment through cathode	(1) class B (3) positive to white light
Trott (1953)	(1) β particles (2) bombardment through cathode	(1) class B (2) positive to red light (3) negative to white light
Trott also reports on the 'activation' of a diamond but in a different sense to Willardson & Danielson (1950).		
Champion & Dale (1956)	(1) β particles (2) not specified	(1) class A (2) positive to red light (3) negative to white light
Urlau (1960)	(1) β particles (2*a*) perpendicular to the applied field	(1) class A (2) positive to red light (3) negative to green light
	(2*b*) bombardment through the anode	(1) class A (2) positive to red light (3) negative to green light
	(2*c*) bombardment through the cathode	(1) class A (2) positive to red light (3) negative to green light
One of the crystals when bombarded through the cathode did not have the class A characteristic. Its response was then negative to red light.		

In addition to the results tabulated, it was shown by Ahearn (1948, 1951), Stratton & Champion (1952) and Champion & Wright (1959) that the counting efficiency of diamonds may vary widely over different portions of the same crystal. A class of dark counting rate/time characteristic not seen in the present work was reported by Urlau (1960). When one of his diamonds was bombarded through the cathode, the decrease due to the space-charge field was not observed. It is believed that this is a special case of our class B_2 characteristic, a hypothesis confirmed by the negative counting response to red light of that diamond under those conditions.

Both Trott (1953) and Willardson & Danielson (1950*b*) have reported on the 'activation' of diamonds.

'Activation' in the sense of Trott corresponds to the development of the class B characteristic.

'Activation' in the sense of Willardson & Danielson is defined in the following way: In some diamonds, illumination with ultraviolet light caused a large negative response. When the light was switched off, the counting increased to a value higher than the previous dark value. This high value of the counting rate was then maintained indefinitely. The counting response to red light of a diamond in the activated state was negative.

These results of Willardson & Danielson are reproducible with our group B_2 diamonds. The activation corresponds to an accelerated development of the class B characteristic.

It is clear that most of the results previously reported by individual authors each working with a few diamonds have also been found in the present work. The results of the present work were obtained under standard conditions—all the diamonds were subjected to similar preparation, field strength, bombardment flux and intensity of illumination. The variety of results is therefore attributable to differences in the diamonds and not to the variability of the experimental conditions.

It was found impossible to distinguish between the diamonds of group A ordinary, group A anomalous and group B on the basis of either their optical absorption or their photoconduction spectra. In terms of these measurements, the diamonds were simply of type Ia as discussed in part I.

A complete analysis should explain in terms of defect structure:

(*a*) the dark characteristics (the classes);

(*b*) the counting response to illumination (the groups);

(*c*) the empirical relation between the dark characteristic of a diamond and its counting response to illumination;

(*d*) the asymmetry of the counting property.

We first interpret the dark characteristics and the counting response to illumination qualitatively.

The magnitude of a charge pulse due to an ionizing particle entering the crystal is given by a Hecht formula (Hofstadter 1948; Urlau 1960)

$$Q = \frac{eN_0W_n}{s}\left[1-\frac{W_n}{s}\left\{1-\exp\left(\frac{-s}{W_n}\right)\right\}\right]+\frac{eN_0W_p}{s}\left[1-\frac{W_p}{s}\left\{1-\exp\left(\frac{-s}{W_p}\right)\right\}\right], \tag{2}$$

where Q is the charge displaced, e is the electronic charge, N_0 the number of electron-hole pairs produced by the incident particle, s the electrode separation and W the mean range of the charge carriers; the subscripts n and p refer to negative and positive charge carriers respectively.

Here W, which governs the pulse height, is given by

$$W = \mu\tau F, \tag{3}$$

where μ is the mobility, τ the lifetime and F the effective field strength. This formula, derived for the case of uniform bombardment perpendicular to the applied field, is called a

Hecht formula because it assumes an exponential decay of the charge carriers, as first proposed by Hecht (1932).

The derivation of the formula also assumes (McKay 1948) that:

(i) the trap distribution is homogeneous throughout the volume of the crystal;

(ii) the immediate recombination of electron-hole pairs is negligible;

(iii) the drift velocity of the charge carriers in the applied field is small compared with the thermal velocity, so that the concept of mobility is valid.

If a number of particles of varying energy bombard the crystal, this will give rise to a spectrum of pulse heights. The height of each pulse is determined by the Hecht formula, with N_0 roughly proportional to the energy of the particle producing it. The counting rate is the sum, taken over unit time, of all the pulses above the threshold. When β particles are counted, as in the present case, there will in general be a number of pulses below the threshold of detection. If the flux and energy distribution of the particles incident on the crystal remain constant, variations in the counting rate may be attributed to variations in the pulse height. If the pulse height is increased, more pulses fall above the threshold of detection, and the counting rate increases.

The charge carriers which in the course of their migration to the electrodes become trapped give rise to a space-charge field opposing the applied field. The existence of the space-charge field is demonstrated by the depolarization counting which is observed when the applied field is switched off. Trapping reduces the electric field and increases the lifetime. Thus both F and τ are variables.

The mobility μ may be also influenced by trapping, which changes the Coulomb scattering, but this effect is small. The space-charge field is comparable with the applied field. This is shown by the initial value of the depolarization counting rate (figure 3 (*a*)). The charge density required to produce a space-charge field comparable with the applied field is about 10^{11} ions/cm^3. Inserting this figure into the Conwell–Weisskopf (1950) formula, we find a contribution to μ^{-1} of 10^{-7} V s cm^{-2}. This is negligible in comparison with the observed μ^{-1} of 10^{-3} V s cm^{-2} (Klick & Maurer 1951; Redfield 1954; Austin & Wolfe 1956; Wedepohl 1957). The theoretical estimate of 10^{11} ions/cm^3 was checked by measuring the photodepolarization current from a polarized counting diamond as a function of the time. The depolarization was excited with ultraviolet light. Integration of this curve yielded a space charge of 6×10^{11} electron charges/cm^3. The effect of charged impurity scattering may therefore be ignored and the mobility regarded as being constant at constant temperature.

It has been shown (Pearlstein & Sutton 1950; Champion & Wright 1959) that the expression (2) gives a very good fit to the curve of observed pulse-height against field.

Our qualitative interpretations of

(*a*) the dark counting rate/time characteristic, and

(*b*) the counting response to illumination

are those obtained from Chynoweth (1951 *a*), Trott (1953), Champion & Dale (1956) and Urlau *et al.* (1961).

(*a*) The development of an initial maximum in the dark characteristic may be understood as follows: The initial increase is due to the filling of traps, causing the lifetimes of charge carriers and therefore the pulse-height to increase.

In class A the subsequent decrease is due to the building up of the space-charge field. The space charge is located in the traps. In the steady state, the lifetimes and effective field strength are both constant.

In class B the space-charge field tends to a limiting value more rapidly than the lifetime increases. The counting therefore passes through a minimum before levelling off to a steady value when the lifetime becomes effectively constant.

The occurrence of the peak D (figure 3(a)), which is sometimes seen when the applied field is switched off, has previously been ascribed to an increase in the lifetimes of the charge carriers. In the present work the magnitude of this peak was found to depend on the magnitude of the previously applied field. Its height decreased as the field strength was increased.

We write

$$\mathbf{F} = \mathbf{E}_A + \mathbf{E}_S, \tag{4}$$

where $\mathbf{E}_A$ is the applied field, $\mathbf{E}_S$ the space-charge field and $\mathbf{F}$ the effective field strength.

The count rate is an increasing function of the effective field strength. In normal counting $\mathbf{E}_S$ opposes F_N and the effective field strength F_N is given by

$$F_N = E_A - E_S. \tag{5}$$

In counting in the absence of an applied field, the effective field strength F_R is given by

$$F_R = E_S. \tag{6}$$

If (i) $E_S < \frac{1}{2}E_A$, then $F_N > E_S$ and the initial reverse count rate is less than the final forward count rate, while if (ii) $E_S > \frac{1}{2}E_A$, then $F_N < E_S$ and the initial reverse count rate is greater than the final forward count rate.

(i) was observed at high field strengths, and

(ii) at low field strengths in a number of counting diamonds.

It was observed (§ 5·1) that whilst most of the pulses during the depolarization counting were of reversed polarity, there were also some forward pulses. We explain this effect as follows:

The distribution of field in a polarized counting diamond is given by Urlau *et al.* (1961). Integrating their formula by parts gives the necessary result that the work done, when unit charge is carried from one electrode to the other, is equal to the voltage applied across the diamond. The field due to the space charge does no work. If the applied potential is removed, the space-charge field remaining is in the reverse direction to the original applied field over a large region near the middle of the crystal, but in the same direction as the original applied field near the electrodes. Hence most of the pulses observed when the crystal counts under its own space-charge field are reversed, but there are some forward pulses as is verified experimentally.

(b) A theory of the counting response to illumination has to account for:

(i) transients in the response;

(ii) transients in the recovery;

(iii) the fact that during illumination the counting rate may be maintained at a steady value different from that in the dark.

Illumination simultaneously increases the effective field strength and decreases the lifetimes of free charge carriers. The increase in the effective field strength comes about through a reduction in the space charge field (Chynoweth 1951 *a*) as follows: When carriers are freed from the traps they drift further towards the electrodes. They may either be trapped at different sites or leave the crystal. The effect is to separate further the centres of the positive and negative space-charge clouds and to reduce their magnitudes, thereby decreasing the space-charge field and increasing the effective field strength. The traps from which the carriers are freed again become available as traps, decreasing the lifetime. When the experimental conditions are altered then one of these factors may change more rapidly than the other, so giving rise to the transients.

All the diamonds have similar optical absorption and photoconduction spectra, that is to say they contain the same defects, as discussed in part I, yet they may exhibit very different counting responses to the same illumination.

We shall explain these differences in terms of differences in the concentrations of the defects.

During the counting experiments free carriers are continuously released by the bombardment. If the trap density is variable from one diamond to another, the rates at which the traps get filled should be proportional to the number empty in unit volume of each crystal. Thus the rate at which space charge accumulates should vary from one crystal to another. Thus in two extreme cases:

(*a*) When the unoccupied trap density is small, the lifetime of the carriers increases strongly, because it is proportional to the reciprocal of the unoccupied trap density. The prediction is the special case of the class B_2 characteristic in which the count rate increases in time to a steady value, with a negative counting response to any light because the lifetime is now the factor most strongly affected.

(*b*) When the unoccupied trap density is large, the lifetime varies slowly and the levels continue to accumulate a space charge until either

(i) the lifetime increases appreciably, or

(ii) the space-charge field compensates part of the applied field and the counting decreases monotonically to a value lower than the initial counting rate; the final observed count rate may be finite or, if the largest pulses are below the threshold, it may be zero.

The prediction of (*b*) (i) is a class B characteristic. Case (*b*) (ii) leads to a class A characteristic with a positive counting response to any light, because the effective field strength is now the most strongly affected factor.

According to this analysis diamonds of class A in which the final steady count rate is zero, or nearly so, show an increase in the count rate under any illumination and are thus of group A anomalous, while those of class B_2 in which the final steady counting rate is greater than the initial show a decrease in count rate under any illumination and are thus of group B. This is the empirical correlation between the classes and the groups.

We now verify analytically that the three-level model can lead to the other observed types of behaviour in which the nature of the responses may depend on the wavelength of the light.

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The model, for the case when there are both electrons in the conduction band and holes in the valence band, is shown in figure 12.

Direct band-to-band recombination of carriers has been shown to be a very slow process in insulators or semiconductors with a large forbidden energy gap (van Roosbroeck & Shockley 1954; Smith 1959, p. 289). Recombination takes place by way of the levels in the forbidden energy gap. This recombination mechanism was proposed by Champion & Wright (1959), Kennedy (1959) and Champion & Kennedy (1965), in order to explain the spread in pulse heights that is observed even when diamonds are bombarded with mono-energetic particles. The mechanism was also proposed specifically for type I counting diamonds by Afanas'eva & Konorova (1964).

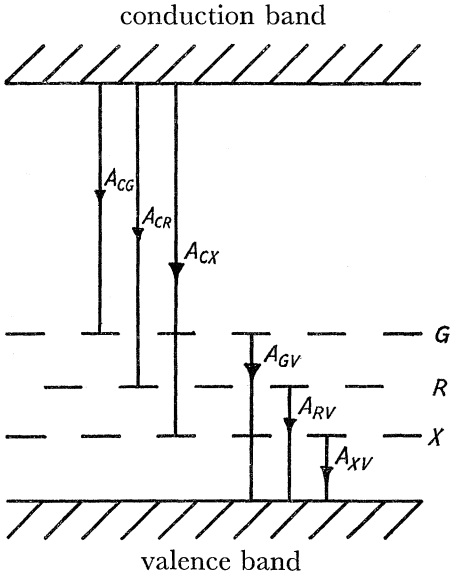

FIGURE 12. The three-level model when there are both electrons in conduction band and holes in the valence band.

We consider in more detail the counting responses under illumination with red light.

For this case, in the absence of bombardment, we write for the level populations, as in § 6 of part I,

$$g_R = g_0+\gamma_R, \quad r_R = r_0+\rho_R, \quad x_R = x_0+\xi_R, \tag{7}$$

where γ_R, ρ_R and ξ_R are the deviations in the populations of the G, R and X-levels during illumination with R-light.

We write

$$\gamma_R = \gamma_{\infty R}+\psi_{GR}(t), \quad \rho_R = \rho_{\infty R}+\psi_{RR}(t), \quad \xi_R = \xi_{\infty R}+\psi_{XR}(t); \tag{8}$$

$\gamma_{\infty R}$ is the value to which the deviation in population of the G-level tends as $t \to \infty$ when the sample is illuminated with R-light. Similarly $\rho_{\infty R}$ and $\xi_{\infty R}$. The ψ's are time dependent, being simply sums of exponentials which tend to zero as $t \to \infty$.

During simultaneous β-bombardment and illumination the populations will in general be different from those given by (7) and (8), but still given by expressions of similar form. We write then

$$g_R = g'_0+\gamma'_{\infty R}+\psi'_{GR}(t), \quad r_R = r'_0+\rho'_{\infty R}+\psi'_{RR}(t), \quad x_R = x'_0+\xi'_{\infty R}+\psi'_{XR}(t); \tag{9}$$

where g'_0, r'_0 and x'_0 are the level populations in the dark steady counting state during a counting experiment.

When the drift distances W_n and W_p are much less than the length of the crystal, s, equation (2) for the pulse height reduces to

$$\begin{aligned} Q &= eN_0(W_n+W_p)/s \\ &= eN_0F(\mu_n\tau_n+\mu_p\tau_p)/s. \end{aligned} \tag{10}$$

Ignoring thermal excitation and direct band-to-band recombination we see that the rates at which the numbers of positive carriers p and negative carriers n are removed from the bands in a crystal in the dark are given by equation (4) of part I

$$\mathrm{d}p/\mathrm{d}t = -A_{XV}xp - A_{RV}rp - A_{GV}gp \tag{11}$$

and by the analogous equation

$$\mathrm{d}n/\mathrm{d}t = -A_{CX}(X-x)\,n - A_{CR}(R-r)\,n - A_{CG}(G-g)\,n, \tag{12}$$

where A_{CX}, A_{CR}, A_{CG} are defined analogously to A_{XV}, etc., in part I.

The reciprocal lifetimes of the charge carriers are given by

$$1/\tau_p = -(1/p)\,\mathrm{d}p/\mathrm{d}t = A_{XV}x + A_{RV}r + A_{GV}g \tag{13}$$

and

$$1/\tau_n = -(1/n)\,\mathrm{d}n/\mathrm{d}t = A_{CX}(X-x) + A_{CR}(R-r) + A_{CG}(G-g). \tag{14}$$

When the crystal is in the dark steady counting state, the reciprocal lifetimes are given by

$$1/\tau_{pD} = A_{GV}g'_0 + A_{RV}r'_0 + A_{XV}x'_0 \tag{15}$$

and

$$1/\tau_{nD} = A_{CG}(G-g'_0) + A_{CR}(R-r'_0) + A_{CX}(X-x'_0). \tag{16}$$

When red light is switched on the reciprocal lifetimes become

$$\begin{aligned} 1/\tau_{pR} &= A_{GV}[g'_0+\gamma'_{\infty R}+\psi'_{GR}(t)] + A_{RV}[r'_0+\rho'_{\infty R}+\psi'_{RR}(t)] + A_{XV}[x'_0+\xi'_{\infty R}+\psi'_{XR}(t)] \\ &= 1/\tau_{pD} + (A_{GV}\gamma'_{\infty R} + A_{RV}\rho'_{\infty R} + A_{XV}\xi'_{\infty R}) + \psi_{pR}, \end{aligned} \tag{17}$$

where

$$\psi_{pR} = A_{GV}\psi'_{GR}(t) + A_{RV}\psi'_{RR}(t) + A_{XV}\psi'_{XR}(t). \tag{18}$$

Similarly

$$1/\tau_{nR} = 1/\tau_{nD} - (A_{CG}\gamma'_{\infty R} + A_{CR}\rho'_{\infty R} + A_{CX}\xi'_{\infty R}) - \psi_{nR}. \tag{19}$$

The terms ψ_{pR} and ψ_{nR} are simply sums of exponentials which tend to zero as $t \to \infty$.

The magnitude of a charge pulse when the crystal is in the steady state is given by

$$Q_D = \frac{eN_0F}{s}\left[\frac{\mu_p}{1/\tau_{pD}} + \frac{\mu_n}{1/\tau_{nD}}\right], \tag{20}$$

and during illumination it is given by

$$\begin{aligned} Q_R &= \frac{eN_0F}{s}\left[\frac{\mu_p}{1/\tau_{pR}} + \frac{\mu_n}{1/\tau_{nR}}\right] \\ &= \frac{eN_0F\mu_p}{s}\left[\frac{1}{\tau_{pD}} + (A_{GV}\gamma'_{\infty R} + A_{RV}\rho'_{\infty R} + A_{XV}\xi'_{\infty R} + \psi_{pR})\right]^{-1} \\ &\quad + \frac{eN_0F\mu_n}{s}\left[\frac{1}{\tau_{nD}} - (A_{CG}\gamma'_{\infty R} + A_{CR}\rho'_{\infty R} + A_{CX}\xi'_{\infty R} + \psi_{nR})\right]^{-1}. \end{aligned} \tag{21}$$

At large times this tends to a steady value which is in general different from that of Q_D. The theory may therefore account qualitatively for both the transient and permanent parts of the response.

To simplify the arithmetic we put $\mu_n = \mu_p = \mu$ (in diamond $\mu_n \sim 1800\,\mathrm{cm}^2\,(\mathrm{V\,s})^{-1}$ (Redfield 1954), $\mu_p \sim 1300\,\mathrm{cm}^2\,(\mathrm{Vs})^{-1}$ (Austin & Wolfe 1956; Wedepohl 1957)), and we assume that the changes effected by the illumination are small, so that (21) may be expanded to first order. We find that at large times the change in the pulse height is given by

$$\begin{aligned} Q &= Q_R - Q_D \\ &\simeq eN_0\mu F\{\tau_{nD}^2(A_{CG}\gamma'_{\infty R}+\ldots) - \tau_{pD}^2(A_{GV}\gamma'_{\infty R}+\ldots)\}/s. \end{aligned} \tag{22}$$

Here τ_{nD} and τ_{pD} are given by (15) and (16), and they depend explicitly on the densities of states of the levels in the forbidden energy gap. Similarly, $\gamma'_{\infty R}$, $\rho'_{\infty R}$ and $\xi'_{\infty R}$ also depend on the defect density, as may be shown from a detailed solution of equations (11) of part I.

The quantity Q may be either positive or negative, and we see from (22) that the change in the pulse height caused by illumination of a counting diamond with red light depends on the densities of states in the levels, their occupancy and their capture cross-sections for the charge carriers of either sign. The latter is the same for all our counting diamonds since, according to their photoconduction spectra, they have the same defects. The former quantities will depend on the relative concentrations of the defects giving rise to the levels, which may vary from one diamond to another. Therefore, depending on the relative concentrations of the defects, the counting diamonds may be divided into two Groups as defined in § 5.2.

Similar results may be obtained when G-light illuminates the crystal.

Because the defects are active both as trapping and as recombination centres, the counting ability of a diamond depends on its purity. In the presence of fast recombination via the defects, the free carriers contributing to a pulse may be rapidly removed. This process accounts for the broad distribution of pulse heights that is observed when counting diamonds are bombarded even with collimated beams of mono-energetic particles (Champion & Wright 1959; Kennedy 1959). The purest diamonds may be expected to be the best counters, whereas in the most impure recombination will be rapid and the pulses will in general be so small as to fall below the threshold of detection. In this respect we are in full agreement with Freeman & van der Velden (1951 *a*).

The theory has up to now assumed that the centres producing the levels in the forbidden energy gap are homogeneously distributed throughout the diamonds.

The asymmetry of the counting described in § 5·1 has its origin in an inhomogeneous distribution of the states. The effect can be accounted for if one type of carrier contributes more to the pulses when the electric field is in a favourable direction than it does when the field is reversed or the crystal rotated through 180°.

Suppose the density of electron trapping sites, δ_n, is roughly constant and the density of hole trapping sites, δ_p, is such that $\delta_p < \delta_n$ at the anode and $\delta_p < \delta_n$ at the cathode.

The contribution of the electrons to the counting rate will be almost the same for the two directions of the applied field. However, if we consider an electron-hole pair produced in a given region of the crystal, one direction of the applied field will cause the hole to move into a region of high trap density, and so to have a short mean free path, while the other field

direction will cause the hole to move into a region of low trap density, and so to have a long mean free path. Thus the total number of pulses above the threshold will be larger for the latter direction of the applied field than for the former. If we had assumed that the distribution of hole traps was uniform, while the distribution of electron traps was non-uniform, this prediction would have been reversed.

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