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1951 Proc. Phys. Soc. A 64 874

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Scintillations from Organic Crystals: Specific Fluorescence and Relative Response to Different Radiations

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MS. received 12th April 1951

ABSTRACT. The scintillation response S of organic crystals depends on the nature and energy E of the incident ionizing particle, of residual range r . The specific fluorescence dS/dr is not in general proportional to the specific energy loss dE/dr . By considering the quenching effect of the molecules damaged by the particle on the 'excitons' produced by it, it is shown that $dS/dr = (A dE/dr)/(1 + kB dE/dr)$. A and kB are constants, which have been evaluated for anthracene from observations of S and E , and the range-energy data. Curves are computed for the relative response S of anthracene to electrons, protons, deuterons and α -particles of E up to 15 mev, and these are shown to agree closely with the available experimental results. The method used for evaluating the relative response is applicable to ionizing particles of any nature or energy, and also to the other organic scintillation crystals

§1. INTRODUCTION

IONIZING radiations impinging on a fluorescent material produce short individual light flashes, or scintillations. These scintillations can be detected with a photo-multiplier tube, and converted into electrical pulses, which can be counted and measured by standard electronic methods. This technique of scintillation counting is being widely applied for the detection and measurement of nuclear radiations. The fluorescent organic crystals are of particular interest for scintillation counting, since they combine a reasonable fluorescent efficiency with a high transparency and a very short luminescent decay time, of the order of 10^{-8} second. These properties make them suitable for the detection of the more penetrating nuclear radiations, and for studies of fast nuclear and meson decay processes, and much of the previous work in this field has been primarily concerned with such applications. The nature of the scintillation process has received rather less attention, and it is, therefore, proposed in this and subsequent papers to consider various fundamental aspects of the fluorescence produced in organic crystals by ionizing radiations.

§2. RESPONSE TO DIFFERENT RADIATIONS

The intensity of the scintillations produced in anthracene and other organic fluorescent crystals depends both on the energy and on the nature of the incident ionizing particle. The amplitude S (volts) of the voltage pulse from a photo-multiplier, operating under constant conditions and observing the crystal, is proportional to the number of fluorescent quanta produced, and hence S may be used as an arbitrary measure of the scintillation intensity. For electrons of energy greater than 125 kev., the scintillation intensity S from an anthracene crystal increases linearly with the energy E (Hopkins 1951), so that the fluorescent efficiency dS/dE (volts/mev.) is constant. For electrons of lower energy, however,

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the efficiency dS/dE is reduced. With heavier particles, which produce more intense ionization, a further reduction in dS/dE occurs, and the scintillation intensity is consequently less than that produced by an electron of the same energy.

It has been noted previously (Birks 1950 a) that the response to different ionizing particles can be readily compared by considering the variation of the specific fluorescence dS/dr (v/cm. air equivalent), which is proportional to the number of fluorescent quanta emitted per unit path length, with the specific energy loss dE/dr (mev/cm. air equivalent), where r (cm. air equivalent) is the residual range of the particle. At low values of dE/dr (electrons of $E > 125$ kev.) the specific fluorescence is proportional to the specific energy loss, corresponding to S increasing linearly with E . At high values of dE/dr (α -particles of $E < 5$ mev.) dS/dr is practically constant and independent of the specific energy loss. The scintillation intensity S is thus proportional to the residual range r of the particle, rather than to its energy E .

§ 3. THEORY

The variation of dS/dr with dE/dr may be readily accounted for on the 'exciton' theory introduced by Bowen *et al.* (1949) to account for the fluorescence of mixed crystals, excited by ultra-violet radiation. On this theory of the energy transfer processes in organic crystals, the electronic energy excited by the incident radiation is transferred from molecule to molecule within the crystal, until it is captured by a single molecule, which then either fluoresces or quenches the 'exciton', depending on the nature of the molecule. This theory has been successfully applied to the energy transfer in mixed crystals of naphthalene and anthracene excited by α -radiation, where both molecular components fluoresce, but the anthracene molecules have the higher 'exciton' capture probability (Birks 1950 b). It has also been used to explain the deterioration of the fluorescent efficiency of anthracene under prolonged α -particle irradiation (Birks and Black 1951). In this case, the molecules damaged by the preceding α -particle irradiation act as quenching agents, having a higher 'exciton' capture probability than the undamaged anthracene molecules.

A similar effect will also occur with individual ionizing particles. The passage of the particle through the crystal produces a local concentration of damaged or ionized molecules along its path. These damaged molecules act as quenching agents for the 'excitons' produced by the ionizing particle. The number of 'excitons' produced per unit path length is proportional to the specific energy loss, say $A dE/dr$. The local concentration of damaged molecules at any point on the particle track is also proportional to the specific energy loss, say $B dE/dr$ molecules per undamaged molecule. Hence, if the 'exciton' capture probability of a damaged molecule, relative to an undamaged molecule, is k , the specific fluorescence

$$\frac{dS}{dr} = \frac{A dE/dr}{1 + kB dE/dr} \quad \dots\dots(1)$$

At low values of dE/dr , (1) becomes $dS/dr = A dE/dr$ and at high values of dE/dr , (1) becomes $dS/dr = A/kB = \text{constant}$, in agreement with the observed behaviour.

The values of A and kB for anthracene have been computed from observations on the relative response to electrons and α -particles reported previously (Birks 1950 a). The residual range r is taken from the range-energy data of Curie (1935) for electrons, and of Livingston and Bethe (1937) and Bethe (1950) for heavier

particles. r is expressed in cm. air equivalent at 15°C. and 760 mm. Hg pressure, but the small correction for the variation of the relative stopping power of anthracene with r has been neglected. This variation is approximately $\pm 1\%$ for $r=0-5$ cm.,

Table 1

| Particles | $E(\text{MeV.})$ | dE/dr | dS/dr |
|---------------------|------------------|---------|---------|
| α -particles | 2.0 | 1.8 | 10.7 |
| Electrons | 0.5 | 0.00215 | 0.175 |

and $\pm 2\%$ for $r=5-180$ cm. The specific energy loss dE/dr is expressed in mev/cm. air equivalent. The experimental values used for substitution in (1) are listed in Table 1, giving for anthracene $A=82.5$, $kB=7.15$.

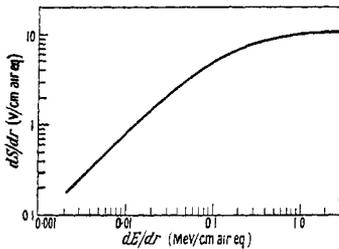


Figure 1. Specific fluorescence dS/dr plotted against specific energy loss dE/dr in anthracene.

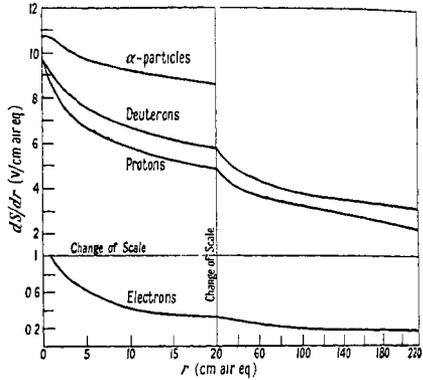


Figure 2. Specific fluorescence dS/dr plotted against residual range r for different particles in anthracene.

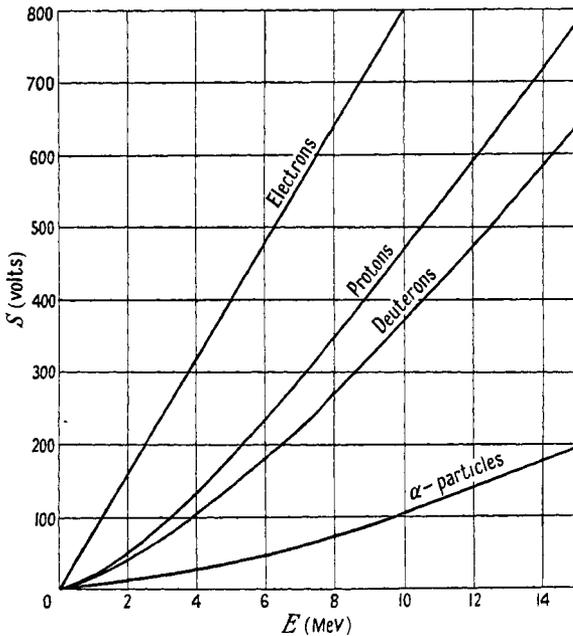


Figure 3. Relative scintillation response S of anthracene to particles of energy E .

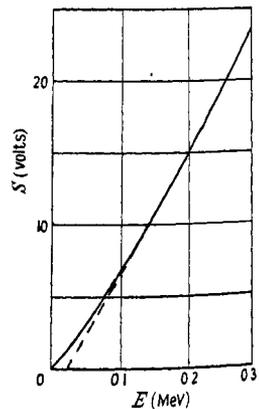


Figure 4. Relative scintillation response S of anthracene to low-energy electrons.

The variation of dS/dr with dE/dr from (1) is plotted in Figure 1. The specific fluorescence dS/dr for different ionizing particles of residual range r has been calculated from (1) and the range-energy data. The results are plotted in Figure 2. The variation of S with r has been obtained by integration of the area below the $(dS/dr, r)$ curves. The scintillation intensity S produced by different ionizing particles of energy E is plotted in Figure 3 and the response to low energy electrons is plotted on an enlarged scale in Figure 4.

§ 4. COMPARISON WITH EXPERIMENTAL DATA

The scintillation response of anthracene to protons of $E \leq 16.4$ mev. has been observed by Franzen *et al.* (1950), to electrons of $E \leq 3.0$ mev. by Hopkins (1951), and to α -particles of $E \leq 5.3$ mev. by Birks (1950 a). In each case the observations agree closely with the predicted responses shown in Figures 3 and 4.

In addition the relative responses of anthracene to different particles have also been reported. A comparison between the experimental response ratios and the predicted values is made in Table 2. The agreement is sufficiently good to establish the general validity of the theory, which may be used to compute the response to any particle.

Table 2

| Reference | Particles | Exptl. ratio | Theor. ratio |
|---------------------------------|---|--------------|--------------|
| Harding <i>et al.</i> (1949) | $\frac{8.78 \text{ Mev. } \alpha\text{-particles}}{6.04 \text{ Mev. } \alpha\text{-particles}}$ | 1.9 | 1.87 |
| | | | |
| Franzen <i>et al.</i> (1950) | $\frac{5.3 \text{ Mev } \alpha\text{-particles}}{0.624 \text{ Mev. electrons}}$ | 0.82 | 0.79 |
| | | | |
| | $\frac{16.4 \text{ Mev protons}}{0.624 \text{ Mev electrons}}$ | 16.4 | 17.5 |
| | | | |
| Hofstadter (1950) | $\frac{5 \text{ Mev protons}}{5 \text{ Mev electrons}}$ | ~ 2.0 | 2.16 |
| | | | |
| Hopkins (1951) | $\frac{5.3 \text{ Mev electrons}}{5.3 \text{ Mev. } \alpha\text{-particles}}$ | 11.6 | 10.8 |
| | | | |

Although the present discussion has been confined to anthracene, for which most experimental data are available, the response curves for other organic crystals to different ionizing particles will be very similar. It is known that the ratio of the response to 5-Mev. α -particles and electrons is approximately the same for anthracene, naphthalene (Birks 1950 a) and terphenyl (Hofstadter 1950), and similar results are to be expected for stilbene, phenanthrene, dibenzyl and the other fluorescent hydrocarbons.

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