tions (6) at certain symmetry points (e.g., $\mathbf{k}=0$) form bases for irreducible representations of the crystal point group, some of which may be automatically orthogonal to core functions made from orbitals of the form (13). At such points we are left with the relatively poor convergence properties of the non-orthogonalized plane waves. For these circumstances Herring has suggested the addition of a Bloch sum of atomic-like functions of the expansion having the appropriate symmetry properties and the proper rapid variation near the nucleus. Callaway has constructed and successfully used such functions in his treatment of iron[14] (if we orthogonalize these functions to the core states as indicated in Part A they do not in principle have to be constructed so as to vanish at half the interatomic spacing as contemplated by Herring and used by Callaway). For our interpolation we first make an initial choice of the vectors $\mathbf{K}_n$ and then for a particular crystal investigate at which points the automatic orthogonality occurs. Then, in order to insure a good representation at and around these points, such atomic-like functions are to be included and treated in the same manner as the core functions.

IV. ACKNOWLEDGMENTS

The author wishes particularly to thank Dr. W. H. Kleiner of the Lincoln Laboratory for many helpful discussions and suggestions.

PHYSICAL REVIEW VOLUME 98, NUMBER 4 MAY 15, 1955

Response of Anthracene and Stilbene to Low-Energy Protons and X-Rays*

J. M. FOWLER† AND C. E. ROOS‡
Department of Physics, The Johns Hopkins University, Baltimore, Maryland
(Received January 20, 1955)

The scintillation response of anthracene and stilbene to low-energy protons (170–570 kev) and x-rays (10–40 kev) has been investigated. In agreement with previous work at higher proton energies, the proton pulse height *vs* energy curves are quite nonlinear. The x-ray data are essentially linear down to 10 kev. This result is contrary to the reported response of these crystals to electron bombardment, but seems to be in agreement with the theory proposed by Birks. With the aid of this theory, proton response curves are obtained for stilbene and anthracene over an extended energy range.

INTRODUCTION

THE response of anthracene and stilbene to protons having energies greater than 500 kev has been extensively investigated.[1–3] Studies have been made of electron response below 500 kev,[4,5] but the response to protons in this energy range has not been reported. It is in this low-energy region that the nature of the scintillation response can be investigated in detail. The work of Hopkins[4] and Taylor *et al.*[3] shows that the response to electrons below 25 kev is nonlinear, and although the proton response is nonlinear above 500 kev, it becomes more pronounced at lower energies.

In the case of low-energy gamma rays or x-rays, the photoelectric process plays the major role. One might therefore expect gamma rays and electrons to have the same response. However, the nonlinearity observed for electrons[3,4] is not apparent in the curves presented here for x-rays which are linear within experimental error. An explanation of this discrepancy can be found in the relation developed by Birks,[6] between the luminescence S and the energy E of the ionizing particle:

$$dS/dr=\phi\frac{AdE/dr}{1+kBdE/dr}. \tag{1}$$

AdE/dr gives the number of "excitons" produced in the crystal by the incident particle. The term "exciton" refers to the quantity which links the energy of the incident particle to the fluorescence. Birks has given arguments which indicate that these "excitons" are photons of about 10-ev energy. The function ϕ is given by the expression

$$\phi=1-\tfrac{1}{2}[\exp(-r/a_0)-(r/a_0)E_i(r/a_0)],$$

where $E_i(r/a_0)$ is the exponential integral, a_0 is the mean free path of the "exciton" in the crystal, and r the range of the incident particle in the crystal. ϕ gives the probability of capture of an "exciton" before it escapes through a crystal surface. $kB\ dE/dr$ represents the quenching of these "excitons" by molecules damaged by the ionizing particle or by inactive impurity molecules.

* This research was supported in part by the U. S. Atomic Energy Commission.
† Now at Washington University, St. Louis, Missouri.
‡ Now at the University of California at Riverside, Riverside, California.
[1] Franzen, Peele, and Sherr, Phys. Rev. **79**, 742 (1950).
[2] Frey, Grim, Preston, and Gray, Phys. Rev. **82**, 372 (1951).
[3] Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).
[4] J. I. Hopkins, Rev. Sci. Instr. **22**, 29 (1951).
[5] J. B. Birks and F. D. Brooks, Phys. Rev. **94**, 1800 (1954).
[6] J. B. Birks, Proc. Phys. Soc. (London) **A64**, 874 (1951).

For photoelectrons produced deep within the crystal, such as is the case with x-rays or high-energy electrons, the probability of an "exciton" escaping is small and ϕ approaches unity. In addition, dE/dr is very small for electrons. For 20-kev electrons dE/dr is 1.70×10^{-2} Mev/cm-air-equivalent, and for 500-kev electrons it is 0.33×10^{-2} Mev/cm-air-equivalent, according to the data reported by Glendenin.[7] Since kB is found to be approximately 6 (see below), the term $kB\ dE/dr$ can be neglected for electrons of energy greater than 500 kev, and it makes only a 10 percent contribution to dS/dr at 20 kev. The formula (1) can therefore be approximated by

$$dS/dE=A, \tag{2}$$

and the curve of S *vs* E is substantially linear for x-rays and fast electrons, which penetrate an appreciable distance into the crystal. For low-energy electrons which do not penetrate very far below the surface, the quantity ϕ is a function of r and therefore of E, and this dependence combined with the $1+kB\ dE/dr$ contribution produces the nonlinear effect which is observed.

For heavy particles such as protons and alphas, where dE/dr is no longer small, there is a significant nonlinearity introduced by the $kB\ dE/dr$ term in (1). In fact, for very highly ionizing particles for which dE/dr is large, (1) approaches

$$dS/dr=(A/kB)\phi. \tag{3}$$

Birks[8] has given values of ϕ plotted against r/a_0. Therefore, using Eqs. (2) and (3), and knowing dS/dE for x-rays and dS/dr for alphas, one can determine A and kB. Then using these values in (1), one can construct curves for dS/dr which are correct for any incident particle. With dE/dr obtained from range-energy curves, this information can then be presented in the more useful form of curves of S *vs* E.

In the following sections, kB and A are calculated from data for low-energy protons and x-rays and the predicted curves of S *vs* E are then drawn for both anthracene and stilbene. The validity of these curves is tested by comparing them with high-energy proton data reported in the literature.

EXPERIMENTAL PROCEDURE

The anthracene and stilbene crystals used in the proton experiment were $1\times1\times0.2$ cm in size, and in the x-ray experiment they were $1\times2\times0.2$ cm. The scintillations were detected with an RCA 6199 photomultiplier tube to which the crystals were directly coupled with optically clear grease. After passing through a linear amplifier, the pulses were analyzed by a single-channel pulse-height discriminator.

The source of protons was an electrostatic accelerator. They were observed after being scattered from a thin (1000 A) nickel foil. A scattered beam was used in order to decrease the danger of radiation damage which is known to result from a high-intensity beam. The proton energy was measured by a generating voltmeter which was calibrated by observing the well-known resonance in the $Li^7(p,\gamma)Be^8$ reaction. The protons were observed at 90° and traveled in an evacuated path from scatterer to crystal. The lowest energies were obtained using the H_2^+ beam of the accelerator.

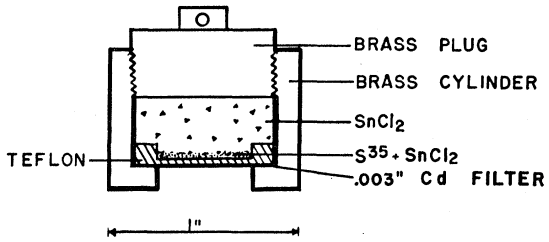

FIG. 1. S^{35}-Sn x-ray source used for calibrations.

In order to study the response of these crystals to electron excitation, monochromatic x-rays of the desired energies were isolated from the general radiation of a tungsten-target x-ray tube by means of a 1.5-meter Bragg single-crystal spectrometer. The regulated high-voltage power supply used in this work has been previously described.[9]

Since the x-ray source and the proton source were in different parts of the laboratory, it was desirable to use an x-ray line as a reference to insure equivalence of gain settings when the equipment was turned off and moved. Several low-energy x-ray sources were constructed by mixing 2 Mc of S^{35}, a pure beta emitter, with the salts of elements such as molybdenum and tin. The details of construction are shown in Fig. 1. The beta rays eject the K electrons from these elements and the characteristic K x-rays are produced. Zirconium and cadmium foils were used to filter out the K_β x-rays.

The S^{35}-Sn source is free of x-rays whose energy is greater than 150 kev and emits very few x-rays above 50 kev. With a filter, the tin K_α lines are approximately 5 times more intense than the general radiation. The peak of the pulse-height distribution obtained from the radiation of the S^{35}-Sn source was found to be the same as that of the monochromatic 25.5-kev radiation obtained from the crystal spectrometer.

RESULTS

Pulse-height distributions from protons incident upon anthracene and stilbene had the expected Gaussian shape with a full width at half-maximum of about 30 percent for 0.46-Mev protons. The pulse heights observed were reproducible to 1 volt, if gain, optics, etc., were kept constant. For a different crystal or a different optical coupling, a relative shift in pulse height was observed, and therefore all proton data reported are for the same anthracene and stilbene crystals.

[7] L. E. Glendenin, Nucleonics **2**, 12 (1948).
[8] J. B. Birks, *Scintillation Counters* (McGraw-Hill Book Company, Inc., New York, 1953), Chap. 6.5.
[9] G. Schwarz and E. H. Byerly, Rev. Sci. Instr. **19**, 273 (1948).

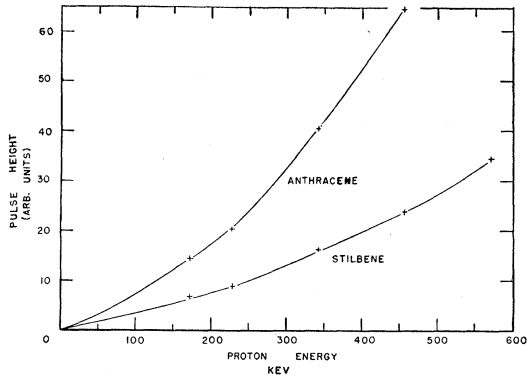

FIG. 2. Pulse heights from anthracene and stilbene as a function of proton energy.

In Fig. 2 is shown the variation of average pulse height with proton energy. The average stilbene to anthracene pulse-height ratio for a common proton energy was 0.41, in good agreement with the value of 0.40 reported elsewhere in the literature for protons of higher energy.[3] The ratio seemed higher at the very low energy points, but in this region the results are very dependent on surface structure, and so can vary from crystal to crystal. The curves are nonlinear in the entire region investigated, in agreement with the relation (1) discussed in the introduction.

The plot of average pulse height *vs* x-ray energy is shown in Fig. 3. With the exception of the 10-kev x-ray points, the mean of each distribution in the pulse height spectrum could be obtained from the axis of symmetry of the distribution. In the case of the 10-kev points, fluorescence pulses obscured the low end of the distributions, making it necessary to determine the mean solely from the location of the maxima of the curves. Wright[10] has recently pointed out that low-energy radiations may give asymmetric pulse-height distributions, making it difficult to assign meaning to the locations of their maxima.

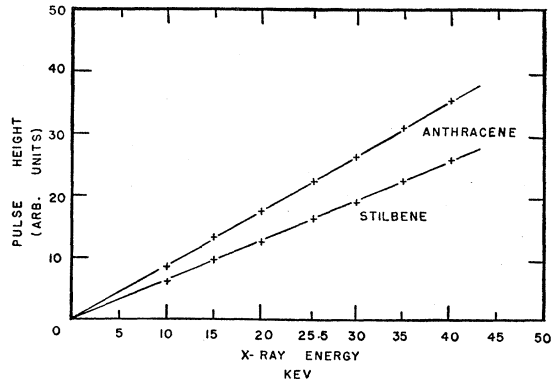

FIG. 3. Pulse heights from anthracene and stilbene as a function of x-ray energy.

[10] G. T. Wright, Phys. Rev. **96**, 569 (1954).

The pulse-height units on the curves in Fig. 3 are the same as those of the proton curves, and this conformity was insured by using the x-ray line from the S^{35}-Sn source as a common normalizing point. The curves are linear within experimental error, and intersect at the point of zero energy. That this point corresponds closely to the point of zero pulse height was confirmed by finding the true zero of the pulse-height analyzer. In terms of the units shown in Fig. 3, the point of intersection of the extrapolated linear curves was found to lie below the origin by 0.5+1.0 units. Hence a slight nonlinearity below 10 kev is not inconsistent with the data. The ratio of pulse height in stilbene to pulse height in anthracene was found to be 0.72. An examination of the data of Taylor *et al.*[3] shows their ratio to be 0.6 approximately.

In Fig. 4 the data are plotted in a manner which is useful in calibrating crystals with natural sources. The energy of the proton is plotted *vs* the energy of that x-ray which produces an equal pulse height. It is seen

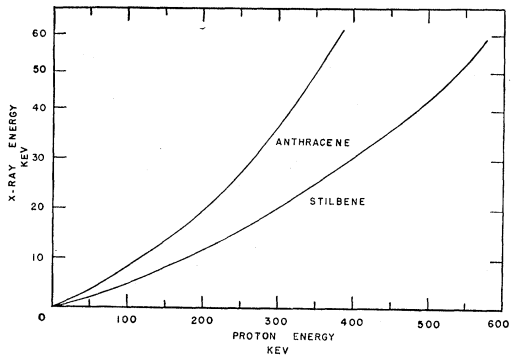

FIG. 4. X-ray energy *versus* proton energy for equal pulse heights.

that in anthracene an x-ray gives approximately the same light output as a proton with ten times its energy, while for stilbene this factor is 15. This factor is not constant, but is larger for the low energies and decreases for higher energies.

DISCUSSION

The relation between the average pulse height and energy observed for x-rays down to 10 kev is compatible with the linear relation (2). Birks and Brooks[5] have recently reported on experiments conducted on anthracene with soft x-rays from secondary radiators. They fitted the results to the theoretical curve predicted by (1) with experimentally determined values of A and kB. A slight nonlinearity is indicated by the theoretical curve but the data seem to be consistent also with a straight line within experimental error. Robinson and Jentschke[11] have also given data for the

[11] W. H. Robinson and W. Jentschke, Phys. Rev. **95**, 1412 (1954).

response of anthracene to x-rays with energies up to 30 kev. The major part of the nonlinearity of response which they reported occurs in extrapolating the measured points to zero energy and zero pulse height.

The quenching factor kB could be calculated directly from dS/dr if one had experimental data from highly ionizing particles such as alphas, where dE/dr is large enough to allow the approximation (3) to be made. For 2-Mev alphas, $dE/dr=1.8$ Mev/cm-air-equivalent[6] and ϕ is approximately one. Birks calculates $kB=7.15$ cm-air-equivalent/Mev on the basis of his experimental value of dS/dr.

The protons in our experiment have a dE/dr of only 0.80 Mev/cm-air-equivalent for the lowest energy (0.174 Mev) and are therefore not highly ionizing enough to use the approximation (3). For a value of $a_0=3$ mm-air-equivalent as given by Birks[5] it is found that ϕ varies from 0.91 to 0.98 in the energy range investigated, and so must be taken into account. kB is

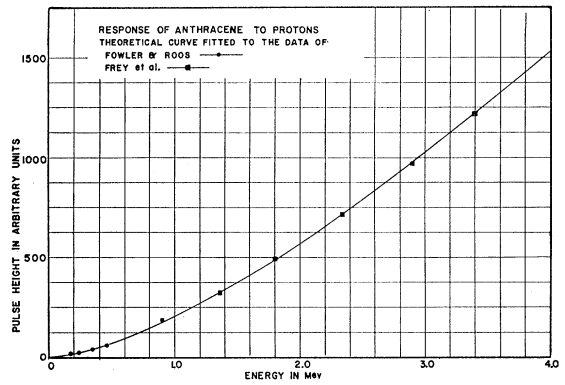

FIG. 5. Pulse heights *versus* proton energy for anthracene. Experimental data of Frey *et al.* (see reference 2) normalized to the curve at 3.4 Mev.

then calculated by using the value of A as determined from the x-ray data, the experimental values of dS/dE, and dE/dr from Bethe's[12] data. Equation (1) is re-

[12] H. A. Bethe, Revs. Modern Phys. **22**, 213 (1950).

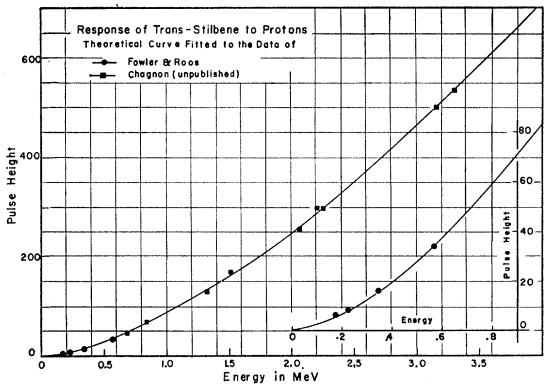

FIG. 6. Pulse heights *versus* proton energy for stilbene. Experimental data of Chagnon normalized to the curve at 3.3 Mev.

written in the form

$$kB=\frac{1}{dE/dr}\left[\frac{\phi A}{dS/dE}-1\right]. \tag{4}$$

The average of the values of kB calculated in this manner are for anthracene, $kB=6.3$ cm-air-equivalent/Mev, and for stilbene, $kB=13.7$ cm-air-equivalent/Mev. The anthracene value is to be compared with 7.15 cm-air-equivalent/Mev reported by Birks.[6] Since kB is the quenching factor which depends upon crystal damage and impurities, the agreement between our value and that of Birks is surprisingly good.

With values of kB and A empirically determined, a theoretical curve of dS/dE was drawn and then integrated to give S *vs* E. This curve is shown for anthracene in Fig. 5 and for stilbene in Fig. 6 for energies up to 4 Mev. To check its conformity with experiment, pulse height *vs* energy data from the literature were normalized at one point and plotted on the theoretical curves. The agreement between experiment and theory is excellent.

The authors would like to thank Dr. J. A. Bearden, Dr. S. S. Hanna, and Dr. G. E. Owen for many helpful discussions, and R. A. Slaven for assistance in the computations. We are also indebted to Paul Chagnon for the proton data above 0.6 Mev in Fig. 6 and the opportunity of using the data before publication.