

Scintillation Response of Phosphors at Low Particle Energies

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The results of a detailed statistical analysis of the scintillation counter are used to correct previously published data for the response to low-energy photoelectrons of NaI(Tl) and anthracene. The nature of the true response curves is discussed.

INTRODUCTION

DEVELOPMENT of the scintillation counter has for the first time permitted quantitative investigation of the luminescence excited in crystals because of individual nuclear particles. Much useful information has been gained in this way about a fundamental aspect of the interaction between nuclear particles and solids.

In this connection it is essential that the characteristics of the scintillation counter are accurately known in order that the observed pulse-height distribution may be satisfactorily related to the scintillation intensity. At moderate and high particle energies the distribution for monoenergetic particles is essentially Gaussian¹ and it is permissible to identify the position of the mean with that of the maximum as generally done. At low particle energies however, the statistical processes of photoelectron production and cascade multiplication of these in the photomultiplier tube result in a distribution which is markedly asymmetrical. A detailed calculation² of these processes has shown that the position of the maximum always lies below that of the mean; for the anthracene scintillation counter this difference becomes significant at electron energies below about 100 kev. It is the purpose of this communication to apply this correction to the previously published data of West, Meyerhof, and Hofstadter³ and Birks and Brooks⁴ for the NaI(Tl) and anthracene counter, respectively.

METHOD

A statistical analysis was made of the experimental distributions in order to convert mean pulse-height measurements \bar{V} , into mean photocathode electrons $\bar{n}\bar{p}$. The general expression for the fractional variance of the observed distribution⁵ is:

$$\frac{\text{var}(V)}{\bar{V}^2} = \frac{1}{\bar{n}\bar{p}} \left(1 + \frac{\epsilon}{\bar{r}} \frac{\bar{s}}{\bar{s}-1} + \frac{\sigma_r^2}{\bar{r}^2} \right) + \text{constant},$$

where \bar{r} is the mean gain of the first dynode stage, \bar{s} is the mean gain per stage of the remaining stages, and σ_r^2/\bar{r}^2 is the fractional variance of the gain of the first stage being due to nonuniform sensitivity over its sur-

face. The quantity ϵ is a statistical enhancement factor, similar to that used by Morton, and is caused by non-normal variance of the dynode gain; under normal operating conditions it may be taken as unity.⁵ The constant is determined by a variety of factors and in a well-designed counter is zero. The fractional variance was derived from the fractional half-width (using only the distributions of Gaussian profile from the higher-energy particles) and plotted in terms of the reciprocal mean pulse height. In each case the resulting curve was linear and passed through the origin. From the slope of this line the original results were interpreted in terms of mean photocathode electrons.

The position of the mean of the distribution differs from that of the maximum np_m . In terms of photocathode electrons we have²:

$$\bar{n}\bar{p} = np_m + \delta,$$

where δ is a constant which is slightly dependent upon multiplier gain but whose average value is about 0.7.

The true response curves are shown in Fig. 1 and Fig. 2 for NaI(Tl) and anthracene, respectively; the original experimental results are also shown in these diagrams to illustrate the appreciable magnitude of the correction. The true response curve for NaI(Tl) indicates that for this material the conversion efficiency increases at very low energies while for anthracene the response is exactly proportional to electron energy.

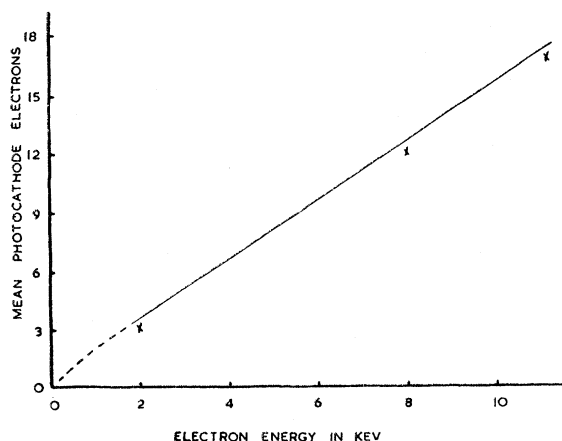


FIG. 1. Response of NaI(Tl) to internally produced photoelectrons. \times experimental; — true response.

¹ G. F. J. Garlick and G. T. Wright, Proc. Phys. Soc. (London) **B65**, 415 (1952).

² G. T. Wright, J. Sci. Instr. (to be published).

³ Mest, Weyerhof, and Hofstadter, Phys. Rev. **81**, 141 (1951).

⁴ J. B. Birks and T. D. Brooks, Phys. Rev. **94**, 1800 (1954).

⁵ G. T. Wright, J. Sci. Instr. (to be published).

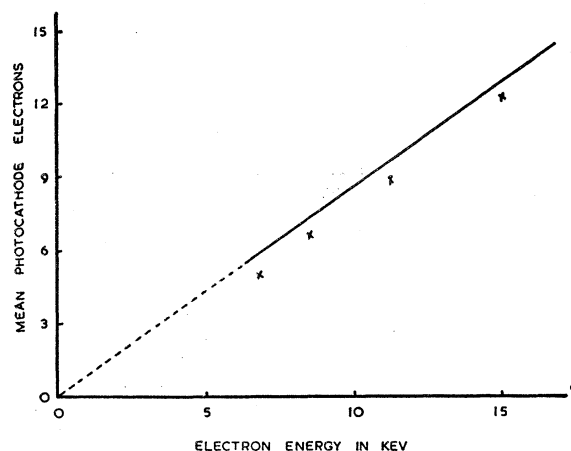


FIG. 2. Response of anthracene to internally produced photoelectrons. \times experimental; — true response.

DISCUSSION

Anthracene is typical of the organic phosphors and these will be discussed as a class.

The nature of the response to internally produced photoelectrons as illustrated in Fig. 2 is of considerable fundamental importance unlike the response curves to external electrons for which surface effects may be appreciable. The true fluorescence intensity is proportional to the total energy dissipated in the crystal over the measured energy range. It is of significance that there appear to be no quenching processes dependent upon nonradiative interaction between ionized or excited molecules for, if present, such mechanisms would produce a nonlinear response. The nonlinear response to external electrons⁶ must consequently be caused wholly by surface effects such as reflection of primary electrons and escape of secondary electrons and soft x-rays (or possibly far ultraviolet photons, see Birks⁷). In addition, overlap of absorption and fluorescence spectra produces considerable variations in scintillation intensity due to a decrease in the efficiency of collection of fluorescence photons as the depth of penetration of the particle decreases. This occurs because fluorescence photons emitted by molecules near the surface may escape through the surface⁸ instead of being reabsorbed in the crystal with possible collection after subsequent re-emission. Measurements by the author (unpublished) show that for short range particles in anthracene ($R < 10^{-3}$ cm) this process can reduce the apparent scintillation efficiency to less than one-half of its actual value.

At very low photoelectron energies, however, one would expect to find a nonproportional response since for energies less than the maximum in the ionization

curve ionization is no longer the predominant mechanism of energy loss and there is probably an associated change in fluorescence efficiency. Measurements in this region would be made most readily by the techniques of vacuum spectroscopy and are likely to yield results of much interest. It is in this region that scintillation decay times become longer than photofluorescence decay times and constant quantum efficiency of response to absorbed photons changes to constant energy response.

Although the response to heavy particles is nonlinear,⁶ the conditions which exist for these are quite different from those for external electrons. For the same initial velocity (which together with charge mainly determines the rate of energy loss), these penetrate many times further through the crystal than electrons and in any event do not suffer the multiple scattering experienced by electrons, which keeps a large proportion near the surface; it is unlikely therefore that surface effects are appreciable for the heavy particles except at very low energies.⁹ The quenching processes operative are consequently to be sought in conditions which are not produced by electrons.

It is known that the heavy particles are proficient in producing damage of the crystal lattice. Each absorbed particle produces a large number of permanently damaged molecules and it is almost certainly the case that many more molecules are temporarily damaged or dissociate but recombine on the Franck-Rabinovitch principle before this damage can become permanent. If we accept the effectiveness of the particle for producing radiation damage as a measure of the extent of this latter process it is negligible for electrons as compared with the heavy particles.¹⁰ It is feasible then that the bimolecular quenching mechanisms¹¹ for the heavy particles are connected with the production of temporary chemical entities in the excitation column which, although recombining rapidly, nevertheless exist for a sufficient period (10^{-11} to 10^{-10} sec) for quenching to occur.

CONCLUSION

The linear response to internally produced electrons shows that quenching by nonradiative interaction of ionized and excited molecules in the excitation column is not the mechanism responsible for the nonlinear response of the organic phosphors to ionizing radiations.

In the case of external electrons this is attributed to surface effects, and for the heavy particles to bimolecular processes connected with the production of temporarily damaged molecules in the excitation column.

ACKNOWLEDGMENTS

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⁶ Taylor, Jentschke, Remley, Eby, and Kruger, *Phys. Rev.* **84**, 1034 (1951).

⁷ J. B. Birks, *Scintillation Counters* (Pergamon Press, London; McGraw-Hill Book Company, Inc., New York, 1953).

⁸ This is not escape of "primary" photons as envisaged by Birks (reference 7).

⁹ G. T. Wright and G. F. J. Garlick, *Brit. J. Appl. Phys.* **5**, 13 (1954).

¹⁰ F. A. Black, *Phil Mag.* **44**, 263 (1953).

¹¹ G. T. Wright, *Phys. Rev.* **91**, 1282 (1953).