Fluorescent Response of CsI(Tl) to Energetic Nitrogen Ions

M. L. HALBERT
Oak Ridge National Laboratory, Oak Ridge, Tennessee
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The light output of a CsI(Tl) crystal was measured as a function of energy for nitrogen ions from 2.9 to 23.8 Mev, for α particles up to 5.30 Mev, and for γ rays of 73, 279.2, and 411.8 kev. The particle energy was varied with absorbing foils. The response to nitrogen ions was proportional to energy up to about 16 Mev. Above this energy a slight curvature of the response curve away from the energy axis was observed. The response to α particles was linear above about 3.5 Mev and extrapolated to $\sim$1 Mev on the energy scale. The light output from the γ rays was proportional to their energy and parallel to the linear portion of the α-particle response. The specific fluorescence dL/dx appears to be an increasing function of the specific energy loss dE/dx.

INTRODUCTION

MEASUREMENTS of the energy response of various phosphors to protons and α particles have shown nonlinearity at low energies.[1–4] This is believed to be a saturation effect, since it occurs at large dE/dx. Ions heavier than α particles have, at a given velocity, a larger dE/dx and so should exhibit this effect more markedly. Aside from the information obtained for the general problem of fluorescence, the heavy-ion response of a crystal is of importance in heavy-ion experiments in which scintillation counters are used as detectors.

For this investigation thallium-activated cesium iodide was chosen mainly because it is not hygroscopic. Once the crystal has been prepared, it is likely to retain its clean surface. Furthermore, there is no need for a moisture barrier over the crystal, in which the energy of the incident particles may be degraded. These considerations are of particular importance in the detection of short-range particles.

The heavy ions used in these measurements were N^{4+} accelerated by the Oak Ridge National Laboratory 63-inch cyclotron. The energy response of CsI(Tl) to low-energy α particles and γ rays from radioactive sources was also studied for comparison.

EXPERIMENTAL METHOD AND RESULTS

The crystal, 0.45 in. in diameter and 0.040 in. thick, was sanded on No. 400 abrasive paper, polished with jewelers' rouge, and then mounted on a microscope slide with Gelva resin. According to a semiquantitative spectroscopic analysis, the thallium content was about 0.1%. Dow-Corning silicone stopcock grease provided the mechanical and optical coupling between the slide and the face of the multiplier phototube, a DuMont type 6291. No reflector was used; the crystal was exposed directly to the incident particles, with the photocathode forming part of the vacuum chamber.

The pulses from the phototube were fed to a cathode follower with a 20-μsec integration time constant, amplified by a Jordan-Bell Al-D linear amplifier with 1-μsec delay-line clipping, and sorted with a Bell-Kelley 20-channel pulse-height analyzer. The linearity of these instruments was established with a pulser and precision attenuator. A test using radioactive sources with a NaI(Tl) crystal on the same phototube showed linear γ-ray response for the entire system for five energies from 33 kev to 1.12 Mev. The data presented below were taken with the pulses attenuated when necessary to stay within this range of pulse heights. It was thus assured that the electronics was always being used in its linear region.

Nitrogen Ions

Nitrogen ions were scattered by a target of gold leaf about 0.18 mg/cm^2 thick which was oriented at 45 deg to the incident beam. A collimator (acceptance angle $\pm$0.7 deg) in front of the crystal selected particles which had been scattered through 90 deg and had passed through the gold. The scattering angle was known to an accuracy of about 3 deg.

The energy (27.9$\pm$0.3 Mev) of the cyclotron beam was determined by measuring the range in nuclear emulsion of protons scattered at zero degree from a hydrogen gas target.[5] Nickel foils were inserted in the incident beam to decrease its energy. The range-energy relation measured by Reynolds, Scott, and Zucker was used to calculate the energy loss of nitrogen ions in nickel.

A weak Po^{210} source placed near the crystal was used as a reference standard. At all energies except the lowest one, the nitrogen spectrum was taken simultaneously with an α spectrum.

The results are given in Table I and Fig. 1; the pulse height of 5.30-Mev α particles is taken equal to 100. Corrections were made for energy loss in the gold and for motion of the center of mass of the nitrogen-gold system. The pulse-height resolution of 7% at maximum energy permitted the determination of the peak pulse height to an accuracy better than 1%. At the lowest

[1] Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).
[2] F. S. Eby and W. K. Jentschke, Phys. Rev. **96**, 911 (1954).
[3] R. H. Lovberg, Phys. Rev. **84**, 852 (1951).
[4] W. T. Link and D. Walker, Proc. Phys. Soc. (London) **A66**, 767 (1953).
[5] Reynolds, Scott, and Zucker, Phys. Rev. **95**, 671 (1954).

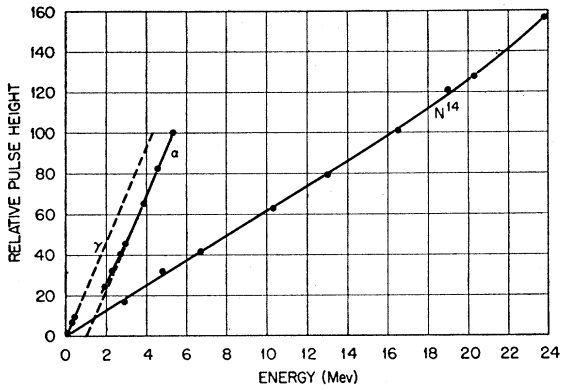

FIG. 1. Response, L, of CsI(Tl) to nitrogen ions, α particles, and γ rays as a function of the energy E, lost in the crystal. The unit of L was fixed by taking $L=100$ for 5.30-Mev α particles.

energy, the estimated error in pulse height was $\pm 3\%$, which arose partly from the poorer resolution ($\sim 35\%$) and partly from a background of low-energy radiation from the cyclotron. The principal errors in the energy scale are due to uncertainties in the scattering angle, the cyclotron beam energy, and the range-energy relation. Each of the first two amounts to about 1%. It is difficult to estimate the error introduced by the range-energy relation. Probably it does not exceed ± 0.75 Mev, as may be judged from the estimated error in the data of reference 5.

Alpha Particles

A second Po^{210} source in vacuum was used for the measurement of α response. The particle energy was varied with aluminum absorbers. Each run at reduced energy was preceded and followed by a calibration run with the previously mentioned weak α source near the crystal. When necessary, corrections for drift in the electronics were made. They usually amounted to less than 0.5% and never exceeded 2.5%. The error in the pulse-height measurement is about 4% at the lowest energy and decreases to $\sim 1\%$ at the full energy.

Conversion of the data to pulse height *vs* energy requires knowledge of the energy loss of α particles in aluminum. As with the nitrogen, the shape of the response curve is sensitive to the choice of the energy-loss relation. The energy-loss function used[6] is based on the experiments of Rosenblum[7] on the slowing of ThC and ThC′ alphas in aluminum. It should be pointed out that the commonly used semiempirical range-energy relations[8,9] do not agree with Rosenblum's data for aluminum.

[6] Landolt-Börnstein, *Zahlenwerte und Funktionen* (Springer-Verlag, Berlin, 1955), 6 Auflage, I Band, 5 Teil, p. 317.
[7] S. Rosenblum, Ann. phys. **10**, 408 (1928).
[8] M. S. Livingston and H. A. Bethe, Revs. Modern Phys. **9**, 271–276 (1937).
[9] Aron, Hoffman, and Williams, University of California Radiation Laboratory Report AECU-663, 1951 (unpublished).

Table I gives the experimental data, and it includes the conversion from absorber thickness to α energy incident on the crystal. The energy could not be calculated for the two thickest absorbers because Rosenblum's data do not go below 2 Mev.

A check on Po^{210} source thickness was attempted with a magnetic spectrometer, but the source was too weak to give a good spectrum. The line profile of a much stronger Po^{210} source of the same area and specific activity indicated a thickness <10 kev. It is therefore likely that the energy loss in the source used for the response measurements was also <10 kev.

After the first series of pulse height *vs* absorber thickness data had been obtained, the crystal was resanded and wiped with a damp tissue, leaving it with a shiny surface. This treatment was intended to remove any nonscintillating layer that might have been present on the crystal. No significant change in pulse height could be detected after the cleaning.

Gamma Rays

Only low-energy γ sources were used with this thin crystal, to insure that most of the photoelectrons would expend all their energy in the CsI(Tl). The results given in Table I are again referred to a standard pulse height of 100 for the 5.30-Mev Po^{210} α particles. The most reliable result, that for the 279.2-kev γ ray from Hg^{203}, is averaged from five measurements, and has an estimated error of about 1.5%. The other two measurements are accurate to about 5%. In Fig. 1 the line determined by the Hg^{203} point and the origin is extrapolated to show clearly that the γ-ray response and the high-energy portion of the α curve are parallel.

DISCUSSION

Nonlinearity in the response of a scintillation crystal is believed to be due to a saturation effect because it usually occurs when the ionizing particle has a large dE/dx. Eby and Jentschke[2] have found that NaI(Tl) becomes nonlinear for dE/dx somewhat larger than

TABLE I. Pulse-height response, L, of CsI(Tl) as a function of E, the energy expended in the crystal. Response to 5.30-Mev α particles is taken equal to 100.

Nitrogen ions			Alpha particles			Gamma rays		
Absorber (mg/cm² Ni)	E[a] (Mev)	L	Absorber (mg/cm² Al)	E[b] (Mev)	L	Source	E (kev)	L
0	23.8	156.1	0	5.30	100.0	Tl x-ray	73	1.79
1.18	20.3	127.3	1.22	4.57	82.1	Hg^{203}	279.2	6.46
1.54	19.0	120.3	2.37	3.84	65.0	Au^{198}	411.8	9.52
2.26	16.5	100.4	3.59	2.94	45.6			
3.18	13.0	78.9	3.95	2.68	40.2			
3.89	10.3	62.5	4.43	2.29	32.0			
4.97	6.7	41.5	4.84	1.95	24.2			
5.62	4.8	31.7	5.20	···	17.4			
6.24	2.9	16.7	5.68	···	9.4			

[a] Based on experimental range curve of Reynolds, Scott, and Zucker.[5]
[b] Based on experimental energy-loss relation of Rosenblum.[6,7]

0.1 Mev/mg-cm^{-2}. Figure 1 shows that in CsI(Tl) the α response curve becomes linear and parallel to the γ-ray response for α energy above about 3.5 Mev. At this energy $dE/dx=0.35$ Mev/mg-cm^{-2}. Thus 0.35 Mev/mg-cm^{-2} is approximately the specific energy loss above which saturation of light output begins. Cesium iodide shows an advantage over sodium iodide in this respect.

The light output, L, of CsI(Tl) to nitrogen ions is linear up to $E\simeq 16$ Mev. An upward curvature becomes noticeable above this energy. It is clear, however, that even below 16 Mev saturation is occurring since dL/dE is only about $\frac{1}{4}$ that of the linear portion of the α response curve. Saturation is to be expected since for nitrogen ions at these energies $dE/dx \gtrsim 2$ Mev/mg-cm^{-2}. It may be that the curvature of the L *vs* E graph continues until the nitrogen passes the energy at which $dE/dx=0.35$ Mev/mg-cm^{-2} ($E\simeq 650$ Mev). Burcham[10] finds, however, that for carbon ions in KI(Tl), L is proportional to E and shows no curvature even up to 113 Mev.

The proton response of CsI(Tl) should be strictly linear at all energies since dE/dx is never greater than about 0.14 Mev/mg-cm^{-2} for protons in CsI.[11] The measurements of Galonsky, Johnson, and Moak[12] for protons from about 0.8 Mev to 4.33 Mev show that the pulse-height response is linear, but extrapolates to 0.07 Mev, which implies that some curvature must exist at low energies. This suggests that either (a) for CsI(Tl) the value of the saturation dE/dx depends on the particle, or (b) at very low proton energies the scintillation process is essentially different from that at high energies. The latter possibility may be connected with the capture of electrons by the proton since this results in a neutral atom which may behave differently from an ion when exciting fluorescence in the crystal.

Comparison of a plot of dL/dx *vs* dE/dx for CsI(Tl) with the corresponding curves by Eby and Jentschke for NaI(Tl) would be of interest. Unfortunately, dE/dx for nitrogen ions in CsI is not accurately known. It has been calculated by (1) obtaining an approximate dE/dx for nitrogen ions in nickel from the slope of the range-energy relation of Reynolds, Scott, and Zucker, and then (2) multiplying by the ratio of the proton stopping powers[11] of Xe and Ni for protons of the same velocity as the nitrogen ions. This procedure is accurate when the particle velocity is large compared with the velocity of the electrons in the stopping medium, but may not be justified at the nitrogen energies considered here. It appears to be satisfactory for nitrogen ions in aluminum.[13] A similar procedure, based on Rosenblum's data for gold,[7] was used to calculate dE/dx for α particles in CsI.

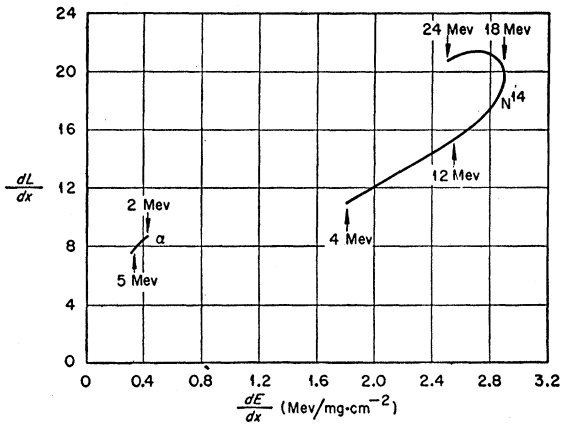

FIG. 2. Specific fluorescence dL/dx of CsI(Tl) as a function of specific energy loss dE/dx for nitrogen ions and α particles. The unit of L is the same as in Fig. 1.

Errors in dE/dx would distort both the ordinate and abscissa of a dL/dx *vs* dE/dx graph. Because of this limitation, the points in Fig. 2 are reliable only for the qualitative behavior of dL/dx. In particular, the fact that dL/dx is double valued may not be significant. It is significant, however, that dL/dx does not decrease at large dE/dx. This behavior is different from that found by Eby and Jentschke for NaI(Tl). The upward trend of dL/dx in CsI(Tl) is confirmed by data of Fulmer and Cohen[14] for fission fragments having dE/dx ranging from 5 to 40 Mev/mg-cm^{-2}.

[10] W. E. Burcham, Proc. Phys. Soc. (London) (to be published).
[11] Taking $Z=54$ for CsI and interpolating from the curves given by S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 793 (1953).
[12] Galonsky, Johnson, and Moak, Rev. Sci. Instr. **27**, 58 (1956).
[13] Webb, Reynolds, and Zucker, Phys. Rev. **102**, 749 (1956).
[14] C. B. Fulmer (to be published).