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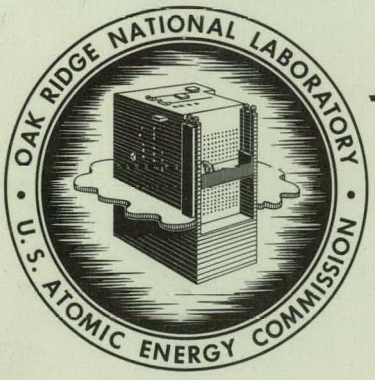
STUDIES OF THE SCINTILLATION PROCESS IN CsI(Tl)

R. Gwin
R. B. Murray



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Neutron Physics Division
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Date Issued

NOV 30 1962

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ABSTRACT

An experimental investigation of the scintillation process in CsI(Tℓ) crystals has been made. The scintillation response of CsI(Tℓ) crystals, having various thallium contents, was measured for excitation of the crystals by monoenergetic gamma rays, protons, and alpha particles. The investigation was made to provide a test of some of the features of a theoretical model of the scintillation process in thallium-activated alkali iodides proposed by Murray and Meyer.

In order to insure that the results obtained in this program would provide a critical test of the scintillation model, special attention was paid to technical effects which could influence the interpretation of the experimental data. For example, the effect of the pulse-analysis time on the relative scintillation response of CsI(Tℓ) to various charged particles was investigated. In addition, the emission spectra of the CsI(Tℓ) crystals were measured for excitation by X rays, protons, and alpha particles. A knowledge of the emission spectra of the crystals is necessary in order to be sure that only the luminescence characteristic of the thallium activator is considered in the analysis of the experimental data.

The results of the investigation have shown that the scintillation efficiency of CsI(Tℓ) is a continuous function of dE/dx, as assumed in the scintillation model, within the accuracy of the experiments when the effect of delta rays are considered. This leads to the conclusion that the light output of CsI(Tℓ) crystals is, in general, a nonlinear function

of the energy of the particle. The shape of the dL/dE versus dE/dx curve for CsI(Tℓ) crystals has been shown, in this work, to be nearly independent of the thallium content of the crystal. This is in distinct contrast to the predictions of the scintillation model.

An examination of the emission spectra of CsI(Tℓ) crystals has shown that light is emitted in a band, centered at about 3300A, which is characteristic of the emission of the pure crystal, as well as the characteristic thallium luminescence. The relative intensity of this luminescence with respect to the thallium-activator luminescence increases as the ionization density in the crystal increases and increases as the thallium content decreases. It has been suggested in this work that the emission band characteristic of pure CsI crystals is associated with the decay of an iodine molecule ion of the type I_2^- which has trapped an electron. On this basis it has been suggested that thallium-activator luminescence results from the successive capture of an electron and a hole at a thallium center. The 3300A emission band of CsI overlaps the optical-excitation spectrum of CsI(Tℓ) for thallium-activator luminescence. Some thallium centers in CsI(Tℓ) are therefore excited by optical emission in the 3300A band.

CHAPTER I

INTRODUCTION

An experimental investigation of the scintillation process in thallium-activated crystals of CsI has been made. The experiments consisted of measuring the relative number of photons emitted by thallium-activated crystals of CsI when irradiated by monoenergetic gamma rays, protons, and alpha particles. The investigation was made in order to test some of the predictions of a theoretical model of the scintillation process in thallium-activated alkali iodides proposed by Murray and Meyer.[1] This model describes the transport of energy deposited in the crystal along the path of a charged particle to luminescence centers formed by the thallium activator.

When energy is absorbed by certain substances, a fraction of the energy may appear as visible or near-visible light. This is the phenomenon of luminescence. If the light appears during the period of excitation, or within about 10^{-8} seconds of the excitation, the process is called fluorescence. The interval of 10^{-8} seconds is the lifetime of an atomic state for an allowed electric dipole transition in the visible region of the spectrum. Light emitted after approximately 10^{-8} seconds is called phosphorescence. The duration of the phosphorescence may be microseconds to hours. When the luminescence is produced by a charged particle traversing the substance, the process is called scintillation.

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[1] R. B. Murray and A. Meyer, Phys. Rev. 122, 815 (1961).

The fundamentals of the scintillation process are not very well understood. The development of photomultiplier tubes led to a considerable effort to find efficient scintillators. Very few attempts have been made, however, to explain the basic mechanism of the scintillation process. An efficient scintillator for use with photomultiplier tubes is one which converts a large fraction of the energy absorbed in the scintillator into light having a wavelength distribution which matches the spectral sensitivity of the photomultiplier tube. Another consideration in the definition of a good scintillator is the duration of the scintillation pulse. If the light from a scintillation pulse is emitted during a period which is long compared to the interval between pulses, the scintillator would not be suitable for most purposes. In the following discussion only phosphors for which the scintillation pulses have a duration of a few microseconds or less will be considered.

During the search for efficient scintillators it was found that pure alkali iodides were good scintillators at the temperature of liquid nitrogen. These same materials were found to be very poor scintillators at room temperature. Pure CsI crystals are particularly good scintillators at -196°C; at this temperature approximately one-half of the energy deposited in a pure CsI crystal by 5-Mev alpha particles appears as photons. It was also found that the addition of certain impurities to the alkali iodides produced scintillators which were reasonably good at room temperature. There is no general agreement on the details of the role played by thallium in forming luminescence centers. The manner in which the energy deposited in a crystal reaches the luminescence centers

is not known, but it is clear that scintillation is not produced by the direct excitation of the luminescence centers by the charged particle. The thallium concentration is orders of magnitude too low to account for the observed photon emission on the basis of direct excitation. About 13 per cent of the energy deposited in thallium-activated NaI by 662-kev gamma rays appears as 3 ev photons.[2] Since it requires approximately 20 ev to produce an electron-hole pair in NaI, the total number of photons emitted in a scintillation event in NaI(Tℓ) is very nearly equal to the total number of electron-hole pairs created in an ionizing event. It is clear that energy absorbed in the crystal must be transported to the luminescence centers. One concept is that the impurity ion is the luminescence center and that it is the source of the light. Johnson and Williams have made calculations of the energy levels of a thallium ion in KCℓ, where the thallium substitutes for potassium in the crystal lattice.[3] The calculations yield an absorption band and an emission band for the thallium ion which are in fair agreement with bands observed in KCℓ(Tℓ). In contrast to this, Knoepfel, Loepfe, and Stoll suggest that their experiments on CsI indicate that the thallium forms a molecular complex with iodine in the crystal and that this complex is the luminescence center.[4] Bonanomi and Rossel stated that the light is emitted by crystalline defects in

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[2]W. J. Van Sciver and L. Bogart, IRE Trans. on Nuclear Sci. NS-5, 90 (1958).

[3]P. D. Johnson and F. E. Williams, Phys. Rev. 117, 964 (1960).

[4]H. Knoepfel, E. Loepfe, and P. Stoll, Helv. Phys. Acta 30, 521 (1957).

the alkali iodides and that thallium only aids in the excitation of the luminescence centers.[5] While the precise role of the thallium in forming a luminescence center is not known, its presence increases the scintillation efficiency of the alkali iodides at room temperature. For optical excitation of KCℓ(Tℓ) crystals the light output is a linear function of the thallium content for crystals having sufficiently low thallium contents.[6] Other elements added to the alkali iodides also produce good scintillators. For example, Tsirlin, Startsev, and Soifer found that the addition of silicon to a CsI crystal produced a good scintillator.[7]

The scintillation model of Murray and Meyer[1] represents an attempt to describe the relative luminescence yield of thallium-activated alkali iodides as a function of the thallium content of the crystal and of the ionization density produced in the crystal by the exciting particle. This model describes the transport of energy deposited by a charged particle in thallium-activated alkali halides to luminescence centers formed by the thallium.[1] Hereafter this model will be referred to as the "scintillation model." The energy carriers are assumed to be a bound electron-hole pair called an exciton. In this model it is assumed that the excitons are formed by the recombination of electrons and holes along the path of

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[5]J. Bonanomi and J. Rossel, Helv. Phys. Acta 25, 725 (1952).

[6]P. D. Johnson and F. E. Williams, J. Chem. Phys. 18, 1477 (1950).

[7]Yu. A. Tsirlin, V. I. Startsev, and L. M. Soifer, Optics and Spectroscopy, 8, 283 (1960).

the charged particle. The excitons diffuse in the crystal until they are captured at thallium-luminescence centers or at other unspecified lattice sites. The scintillation model describes the migration of excitons with a one-velocity diffusion equation under the assumption that in a single scintillation event a thallium center can capture only one exciton. The density of thallium centers which have not captured an exciton is thus a function of space and time. To describe this phenomenon a second differential equation giving the time dependence of the density of the thallium centers which have not captured an exciton was coupled with the diffusion equation. The density of traps was assumed to remain constant during a scintillation event.

The scintillation model was formulated in terms of differential quantities. As the charged particle traverses the crystal, it loses an amount of energy ΔE in a distance Δx along its path. The unit of length Δx in this work refers to an areal density (mg/cm^2). The linear density of electrons and holes along the path of the particle was assumed to be proportional to the differential energy loss dE/dx of the particle in the crystal. The stopping power is defined as $-dE/dx$, where x is in units of mg/cm^2; dE/dx will be used in units of $kev\text{-}cm^2/mg$. The stopping power is a slowly varying function of the energy of the particle, and on this basis, the assumption was made that excitons formed along the length of path Δx diffuse in a cylindrical slice of the crystal of thickness Δx. Diffusion out of the slice was assumed to be balanced by diffusion into the slice from adjacent slices. The excitons captured by thallium-luminescence centers produce a number of photons ΔL from the energy ΔE

deposited along Δx. The model yields the scintillation efficiency, defined as the slope of the light output versus energy curve dL/dE, as a continuous function of the differential energy loss dE/dx. This leads to the conclusion that the light output is, in general, a nonlinear function of the energy of the exciting particle.

Figure 1 shows the calculated scintillation efficiency as a function of the differential energy loss for a CsI crystal having a particular thallium content.* The experimental data available to Murray and Meyer[1] are also shown.** The initial rise of the scintillation efficiency at low values of dE/dx is described by the model in terms of the formation of excitons by the recombination of electrons and holes. The probability that electrons and holes combine to form excitons is proportional to the electron and hole density and these densities are taken to be proportional to dE/dx. This results in the density of energy carriers being an increasing function of dE/dx. The falling scintillation efficiency at large values of dE/dx is accounted for in the scintillation model by the depletion of thallium luminescence sites which have not captured an exciton during a given scintillation event. This feature of the scintillation model leads to the prediction that the shape of the scintillation efficiency as a function of the stopping power is dependent upon the thallium concentration. The scintillation model represents the first

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*Figure 31 shows the calculated scintillation efficiency as a function of dE/dx for crystals having different thallium contents.

**The experimental data used in the formulation of the scintillation model are documented in Ref. 1.

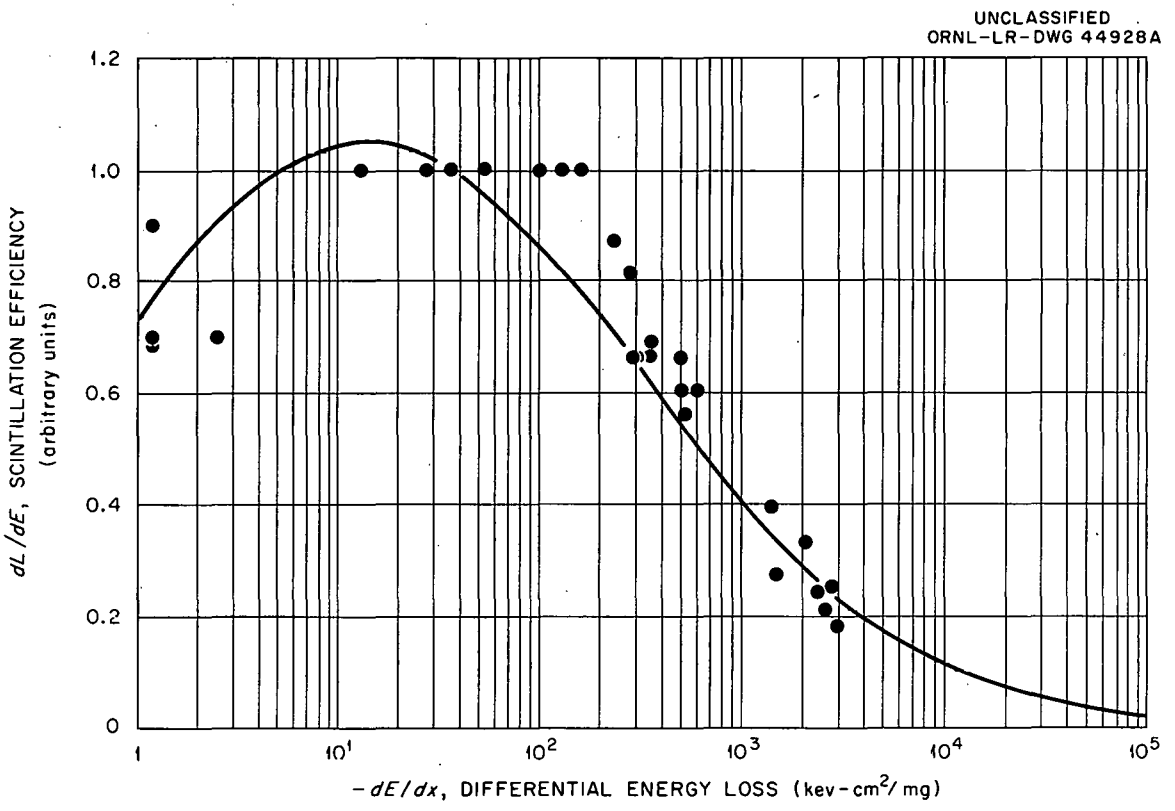

Fig. 1. Calculated Scintillation Efficiency of CsI(Tℓ) Versus dE/dx. The points represent the data on CsI(Tℓ) available for the formulation of the scintillation model.

attempt to explain the observed behavior of the scintillation efficiency of the alkali halides for charged particles in a quantative way.

The data used in the formulation of the scintillation model were obtained from many sources in the published literature. Two factors may introduce considerable uncertainty in the normalization of one set of experimental data to another: (1) the thallium concentration of the crystals used in the various experiments was surely not the same, and (2) the scintillation pulse-analysis time was not the same in all cases. A knowledge of the thallium concentration of the crystals is needed because it is a parameter of the scintillation model. Since the scintillation pulse does not have the same time dependence for excitation by different particles, the relative light output for different particles is a function of the pulse-analysis time.[8,9,10] The scintillation model yields the density of thallium sites which captured an exciton in a scintillation event. The number of photons emitted by the thallium-luminescence centers in a scintillation event is assumed to be proportional to the number of excited thallium centers. In order to interpret experiments on the scintillation process with respect to the scintillation model, it is necessary to consider some of the fundamentals of the scintillation process.

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[8]F. S. Eby and W. K. Jentschke, Phys. Rev. 96, 911 (1954).

[9]R. S. Storey, W. Jack, and A. Ward, Proc. Phys. Soc. LXXII, 1 (1958).

[10]J. C. Robertson and J. G. Lynch, Proc. Phys. Soc. LXXVII, 751 (1961).

The emission spectra of CsI(Tℓ) and NaI(Tℓ) crystals change as thallium is added to the crystals.[11,12] The intensity of emission bands which are emitted in the pure materials decreases as thallium is added to the system while the emission intensity of the band due to the activator increases. The emission spectra of CsI(Tℓ) and NaI(Tℓ) crystals also vary with the exciting radiation;[11,13] this phenomenon is more pronounced at low thallium concentrations. The emission spectrum of a pure CsI crystal depends upon the method by which the crystal was grown.[14]

Van Sciver measured the emission spectra of NaI(Tℓ) crystals for various amounts of thallium.[11] There was a prominent band at 4300A due to the addition of thallium. For pure NaI two bands were observed, one at 3100A and the other at 4100A. The 4100A band was more intense for gamma-ray excitation than for alpha-particle excitation. Van Sciver proposed that the luminescence centers associated with the 3100A and the 4100A emission bands are excited by different mechanisms.[11] In one case the luminescence center is excited by the successive capture of an electron and a hole (4100A band); in the other case electrons and holes which have recombined to form an excited state of the iodine, called an exciton, undergo a radiative transition and the band at 3100A results. The emission spectrum characteristic of the thallium-activator luminescence

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[11]Wesley Van Sciver, IRE Trans. on Nuclear Sci., NS-3, 39 (1956).

[12]Z. L. Morgenshtern, Optics and Spectroscopy, 7, 146 (1959).

[13]G. Hrehuss, Nucl. Inst. and Methods 8, 344 (1960).

[14]Z. L. Morgenshtern, Optics and Spectroscopy 8, 355 (1960).

was the same for excitation of the crystal by photons having wavelengths in the thallium-absorption band of the crystal or for excitation by charged particles. For this reason, energy carriers were proposed which would return the thallium center to its ground state. Van Sciver proposed that the fraction of electrons and holes which recombine to form excitons is proportional to the ionization density produced by the radiation.[11] The ratio of excitons to free electrons and holes would be greater for excitation by alpha particles than for excitation by gamma rays; consequently the 3100A band would be relatively greater for alpha-particle excitation than for gamma-ray excitation. One other possible explanation of this effect may be that one of the luminescence centers is saturating. As the density of energy carriers increases, the density of non-excited luminescence centers of one type may decrease with respect to luminescence centers of another type. Further increases in the energy-carrier density then would excite relatively few luminescence centers of one kind. The relative intensity of the two emission bands would then depend upon the ionization density in the crystal.

Another item which must be considered in analyzing experimental data on the scintillation response of the thallium-activated alkali iodides is the time dependence of the scintillation pulse. In the thallium-activated alkali iodides the rate of emission of photons in a scintillation event rises to a maximum in a time which is short compared to the duration of the scintillation pulse. The emission rate then decreases such that one-half or more of the total light is emitted in one or two microseconds. The time dependence of the scintillation pulse

in thallium-activated alkali iodides has been found to be dependent upon the ionization density produced in the crystal by the charged particle.[8,9,10]

Storey, Jack, and Ward measured the time dependence of the scintillation pulse from CsI(Tℓ) and found that the light intensity I could be represented by the relation[9]

$$I = A_1 e^{-t/\tau_1} + A_2 e^{-t/\tau_2}. \qquad (1)$$

The relative values of A_1 and A_2 and the value of τ_1 were found to be dependent upon the identity of the particle exciting the crystal. The value of τ_1 ranged from 0.43 μsec for excitation by 4.8-Mev alpha particles to 0.70 μsec for excitation by 662-kev gamma rays. The value of τ_2 was 7 μsec for excitation by all particles. Storey, Jack, and Ward stated that τ_1 seemed to be a continuous function of the ionization density produced in the crystal by the charged particle.[9] The number of photons L(t) emitted in the scintillation pulse from its beginning to time t is given by the integral of Equation (1) as

$$L(t) = A_1 \tau_1(1 - e^{-t/\tau_1}) + A_2 \tau_2(1 - e^{-t/\tau_2}). \qquad (2)$$

The fraction of the total light emitted in the first t μsec of the scintillation pulse is given by the relation

$$\frac{L(t)}{L(\infty)} = \frac{(1 - e^{-t/\tau_1}) + \frac{A_2\tau_2}{A_1\tau_1}(1 - e^{-t/\tau_2})}{1 + \frac{A_2\tau_2}{A_1\tau_1}} . \qquad (3)$$

Since the ratio of A_1 to A_2 varies and τ_1 varies with the exciting particle, the ratio $L(t)/L(\infty)$ also depends upon the exciting particle. It is clear that the number of photons emitted during a fixed interval of time, after the start of the scintillation pulse, is not a constant fraction of the total number of photons emitted in the pulse. The time, measured from the start of the scintillation pulse, during which the photons are measured is called the pulse-analysis time. The number of photons collected for a pulse-analysis time t will be referred to as $L(t)$.

Sastry and Thosar measured the time dependence of the scintillation pulse from CsI(Tℓ) in the blue region and the yellow region of the emission spectrum in separate experiments.[15] The object of the experiments was to determine whether the broad emission spectrum and the time dependence of the scintillation pulse from CsI(Tℓ) might be due to two separate emission bands having different transition probabilities. The experiments showed that the time dependence of the scintillation pulse was the same, within experimental uncertainties, for the blue and yellow region of the emission spectrum.

The purpose of the present work is to provide experimental data on the scintillation process in CsI(Tℓ) which will provide a critical test

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[15]N. P. Sastry and B. V. Thosar, Proc. Indian Acad. Sci. 54A, 140 (1961).

of the scintillation model. CsI(Tℓ) was chosen because it is a relatively good scintillator and is not hygroscopic. In addition, there was more information available on the time dependence of the scintillation pulse from CsI(Tℓ) than for the other thallium-activated alkali iodides.[9] The experimental program consisted of measuring the light output of CsI(Tℓ) crystals for excitation by monoenergetic gamma rays, protons, and alpha particles. Several CsI(Tℓ) crystals having thallium contents ranging from 0.002 to 0.31 mole per cent were used in the experimental program. The range of stopping power covered in these experiments was from about 2.5- to 640-kev-cm^2/mg; this range is sufficient to provide a check on the assumption of the model that the scintillation efficiency is a continuous function of the stopping power. The range of the thallium content of the crystals used provides a check on the predicted dependence of the scintillation efficiency on the thallium concentration of the crystals.

The emission spectra of the crystals used in the experiments were measured for alpha particles, protons, and X rays. The measurements extended from about 3000A to 7500A. These measurements were made in order to correct the experimental data for the wavelength dependence of the light detector.

The variation of the time dependence of the scintillation pulse from excitation by different particles was considered in the design and interpretation of the experiments. The scintillation pulse was analyzed at both seven microseconds and one microsecond. On the basis of the experiments by Storey, Jack, and Ward, the seven-microsecond analysis

results in the collection of at least 80 per cent of the total light in the scintillation pulse.[9]

The scintillation experiments performed during the present work do provide a critical test of some features of the scintillation model. The particular features which have been investigated are:

1. The continuity of dL/dE versus dE/dx has been investigated for different particles having nearly the same dE/dx.
2. The monotonic dependence of dL/dE on dE/dx at large values of dE/dx has been investigated. The light output per unit energy for alpha particles has been measured for an energy range where dE/dx passes through a maximum. For this behavior of dE/dx the light output is expected to pass through a minimum.
3. The dependence of dL/dE versus dE/dx upon the thallium concentration of CsI(Tℓ) crystals has been investigated. The range of thallium contents of the crystals used was such that a meaningful test of the scintillation model can be made.

CHAPTER II

EXPERIMENTAL METHODS

I. PREPARATION OF THE CsI(Tℓ) CRYSTALS

The CsI(Tℓ) crystals used in this experimental program were grown by the Stockbarger method. Two of the crystals used were grown in an available furnace at the Oak Ridge National Laboratory. The other crystals were obtained from the Harshaw Chemical Company.

The CsI used to grow the two crystals was obtained from A. D. McKay, Inc. A spectrographic analysis showed that the CsI was essentially free of other alkali metals. The concentration of potassium was about 0.02 per cent by weight and smaller concentrations of other alkali metals were present. There were faint traces of other elements in the CsI; silver, magnesium, iron, and silicon were present in concentrations of the order of 0.0005 per cent.

The crystal-growing apparatus is shown in Figure 2. Powdered CsI was mixed with thallium iodide and placed in a quartz-lined Vycor crucible. The long neck of the crucible was secured to a piston, as shown in Figure 2, which regulated the motion of the crucible. The rate of fall of the piston was controlled by the flow of oil out of the cylinder and through a sintered glass filter. The crucible was positioned near the top of the furnace and connected to a vacuum system. The crucible was maintained at a temperature of about 350°C and held at a reduced pressure for several hours to insure that no moisture remained in the mixture.

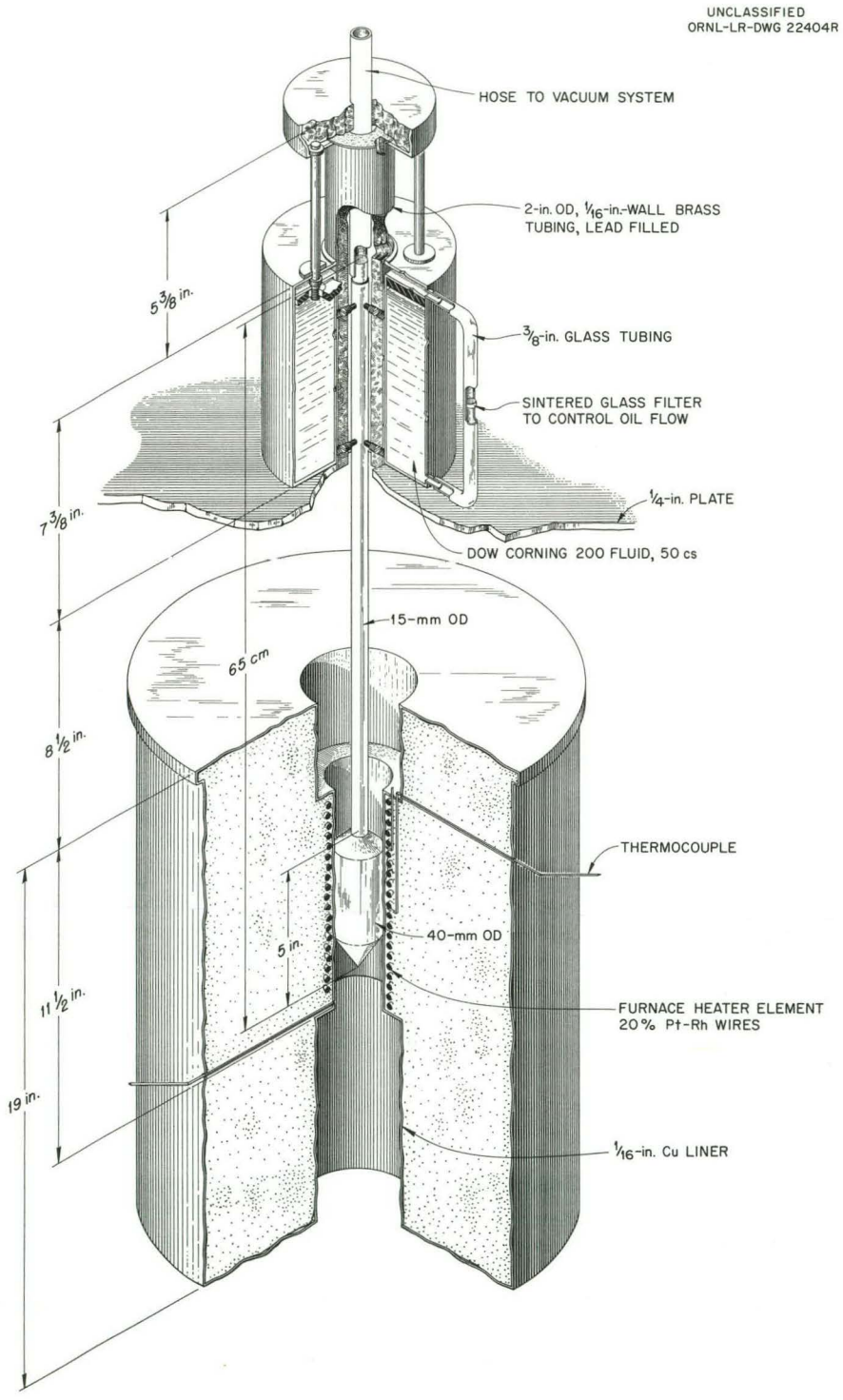

Fig. 2. Apparatus for Growing Single Crystals

The crucible was then filled with helium to a pressure of about one atmosphere and sealed. The temperature of the furnace was increased to about 650°C at the control point in order to melt the cesium iodide and the thallium iodide. The melting point of cesium iodide is 621°C; that of thallium iodide is 440°C. The crucible was then allowed to fall through the furnace which was designed to have a sharp temperature gradient at one point. As the melted mixture of cesium iodide and thallium iodide passes through this temperature gradient, crystal growth occurs. The point on the bottom of the crucible facilitates the growth of a single crystal. After passing through the furnace, the CsI(Tℓ) crystal was brought to room temperature in a twenty-four hour cooling period in order to minimize the introduction of defects in the crystal. During the cooling process the thin quartz liner breaks away from the walls of the crucible as the CsI(Tℓ) crystal contracts. Without the quartz liner the crystal adheres to the walls of the crucible which breaks on cooling, and large cracks and strains occur in the crystal.

The ingots of CsI(Tℓ) grown in the quartz-lined crucibles were about 4 cm in diameter and 5 cm long. The ingots of CsI(Tℓ) obtained from the Harshaw Chemical Company were of varying sizes. The CsI(Tℓ) crystals used in the experiments were cut from the various ingots to have a diameter of 2.5 cm and a thickness of 0.2 cm. The thallium content of the crystals was determined by the method of polarographic analysis; a sample of the ingot of CsI(Tℓ) adjacent to the slice cut for the

scintillation experiment was used for the chemical analysis.*

The surfaces of the crystal were prepared each time the photomultiplier system was assembled. A tissue paper, sprinkled with zinc oxide, moistened with methyl alcohol, and stretched over a flat surface, was used to polish the crystals. An area of the tissue paper was left free of zinc oxide so that as the crystal was rotated by hand on the tissue paper, the zinc oxide could be wiped off the crystal. Crystals treated in this manner had a smooth, clear surface.

II. DESCRIPTION OF THE PHOTOMULTIPLIER ASSEMBLY

The light from the scintillation pulse was detected by an RCA C-7261 photomultiplier tube, having an S-20 response, which is now available as the RCA-7326. The maximum sensitivity of this tube occurs at a wavelength of about 4200A; at 5700A, where the peak intensity of the CsI(Tℓ) emission spectrum occurs, the sensitivity is about one-half the maximum value. The spectral sensitivity of this tube at 7000A is about 20 per cent of the maximum sensitivity. Figure 3 shows the relative spectral sensitivity characteristic of the RCA-7326 photomultiplier tube.** A drawing of the photomultiplier assembly is shown in Figure 4. The crystal was coupled to the face of the photomultiplier tube by a Lucite light pipe which was about 3.8-cm long and 3.8 cm in diameter; the

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*The chemical analysis of the CsI(Tℓ) crystals was performed by D. A. Costanzo and H. Kubota of the Oak Ridge National Laboratory.

**The values of the spectral sensitivity of the RCA C-7261 photomultiplier tube were taken from the specifications supplied by the Radio Corporation of America.

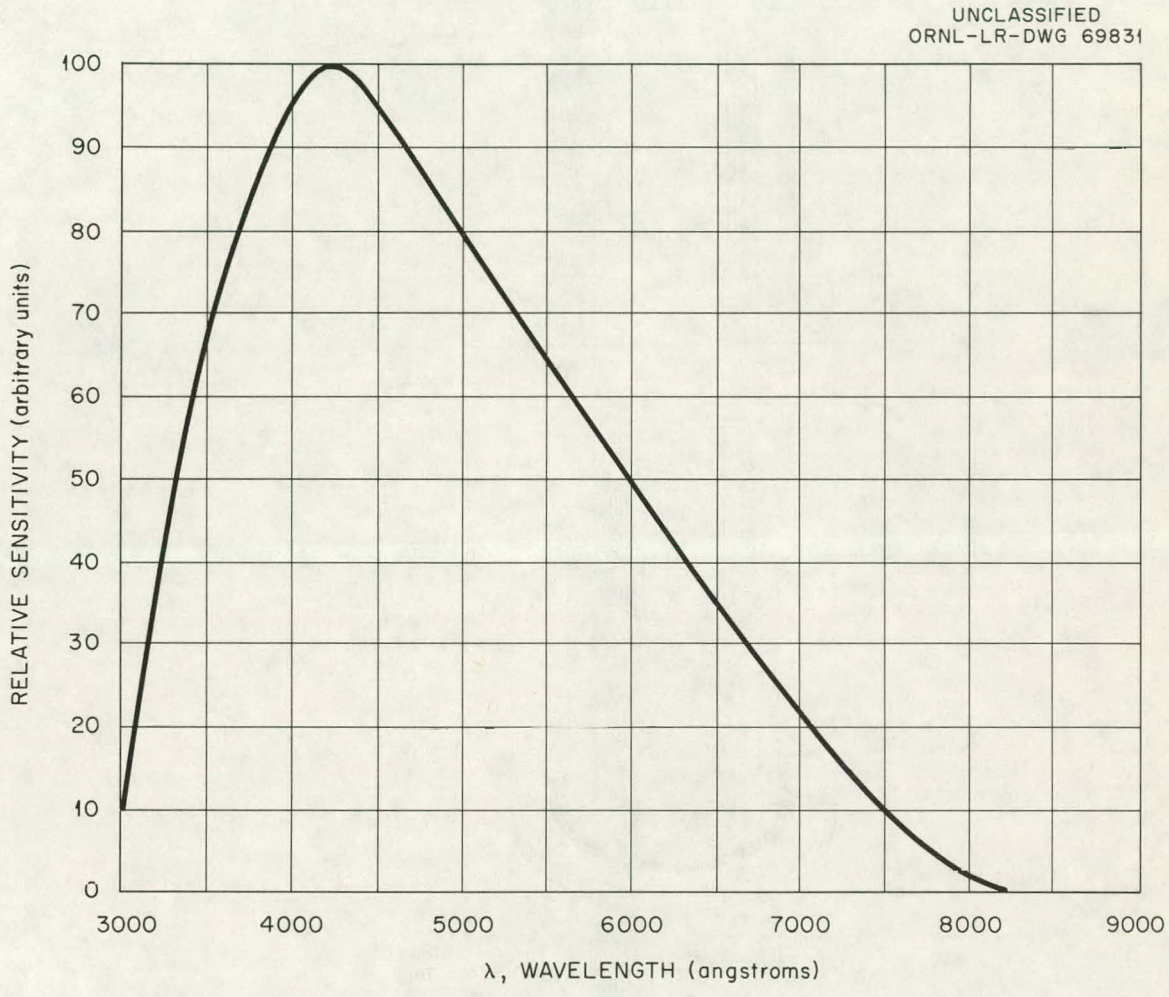

Fig. 3. Typical Spectral Sensitivity of a Photomultiplier Tube Having an S-20 Response

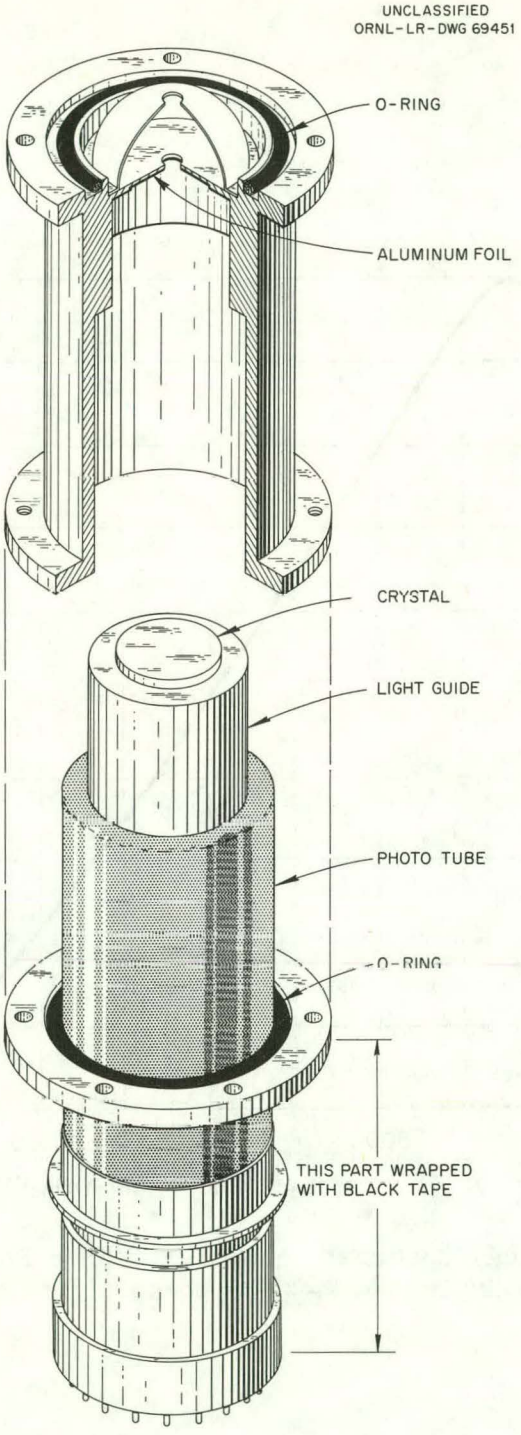

Fig. 4. Diagram of the Photomultiplier Assembly Used in the Scintillation Experiments in Which the Crystal Was Excited by Charged Particles

cylindrical surface of the light guide was wrapped with aluminum foil. The substance used to couple the crystal optically to the light pipe and the light pipe to the photomultiplier tube was Dow-Corning Silicone Q C-2-0057. The aluminum housing, shown in Figure 4, was designed to be used in the vacuum system of a Van de Graaff accelerator and to hold the photomultiplier tube, light pipe, and crystal in a fixed position. The top of the crystal was covered with aluminum foil except for a 0.32-cm-diameter circle in the center. The base of an aluminum hemispherical shell, having a 0.32-cm-diameter hole in the top, was positioned at the top surface of the crystal. The inside surface of the hemispherical shell was highly polished in order to reflect light from the scintillation pulse back into the crystal. The beam of charged particles passed through the hole in the hemispherical shell and struck the crystal. A photograph of the photomultiplier assembly is shown in Figure 5.

The photomultiplier assembly used for the gamma-ray experiments was similar to the one used for experiments with charged particles except that aluminum foil covered the entire top surface of the crystal.

The high-voltage power supply for the photomultiplier tube was a model N-401 of the Hamner Electronics Company. The power for the high-voltage supply was regulated by a constant voltage transformer. The high-voltage output was continuously monitored during the experiments; voltage changes which would have changed the gain of the photomultiplier tube by as much as 0.1 per cent could be detected.

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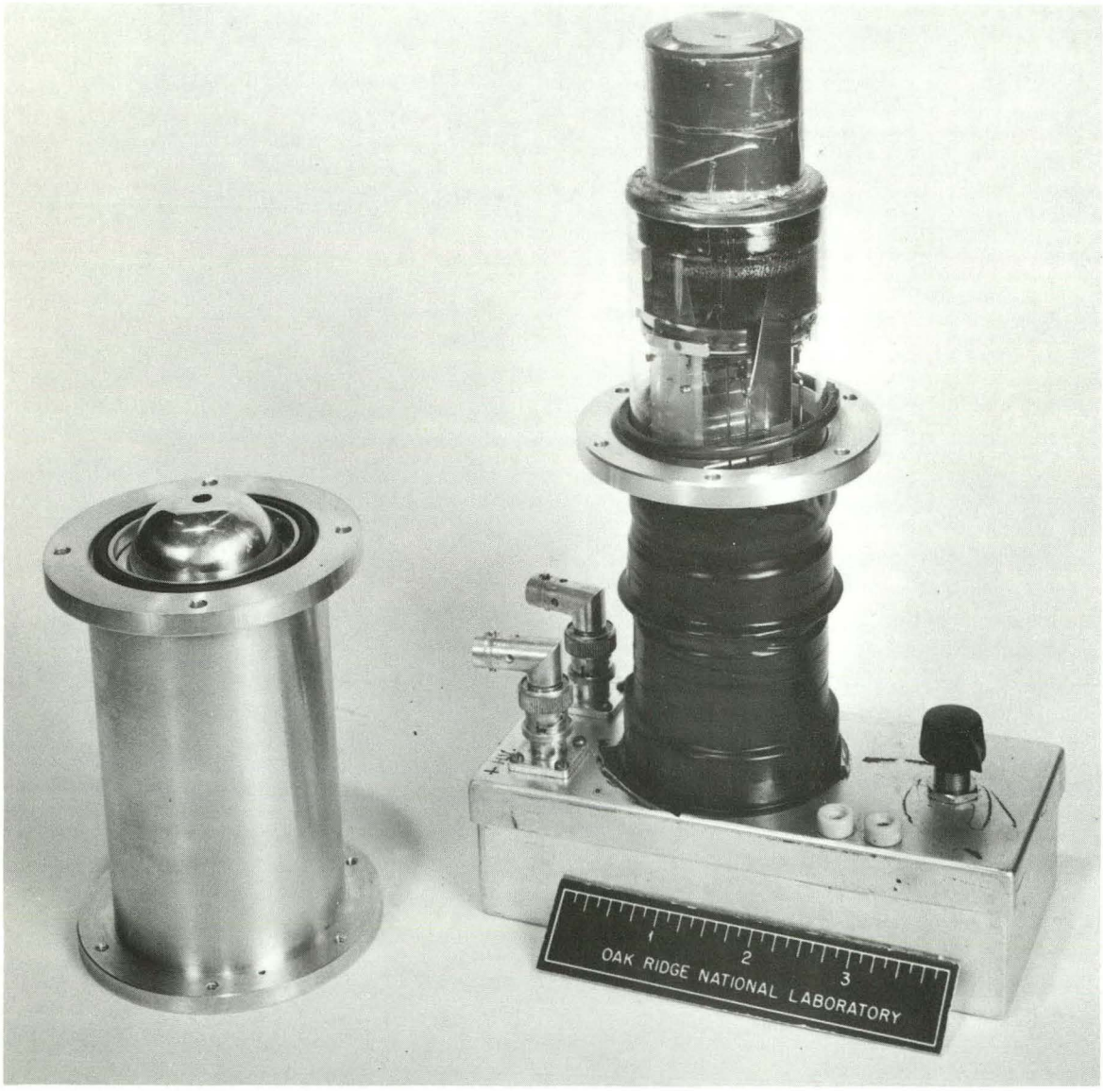

Fig. 5. Photograph of the Photomultiplier Assembly Used in the Scintillation Experiments in Which the Crystal Was Excited by Charged Particles

III. DESCRIPTION OF THE PULSE AMPLIFICATION AND ANALYZING SYSTEM

The current pulses from the photomultiplier tube were integrated at the anode of the tube by a condenser and resistor, in parallel, which gave a time constant of about 250 microseconds. A diagram of the integrating circuit used and the shape of the voltage pulse resulting from integrating a current pulse having an exponential form are shown in Figure 6.

The voltage pulses from the integrating circuit at the anode of the photomultiplier tube were introduced into the preamplifier of a linear amplifier in which the pulses were clipped by double delay lines. The output pulses of the amplifier were analyzed by a multichannel pulse-height analyzer.

Two separate amplifying and pulse-analyzing systems were used in the experiments, one for a pulse-analysis time of seven microseconds and the other for a pulse-analysis time of one microsecond. A modified DD-2 amplifier was used for the pulse-analysis time of seven microseconds. The delay lines used to obtain the seven-microsecond clipping time were made from HH-2000 cable. An A-8 amplifier was used for the pulse-analysis time of one microsecond. The multichannel analyzer used for the pulse-analysis time of seven microseconds was an Atomic Instrument Company 120-channel analyzer; twenty channels could be scanned at a time. This analyzer was modified by the addition of a long-pulse adapter which permitted the use of the seven-microsecond pulse-clipping time. A dual input oscilloscope

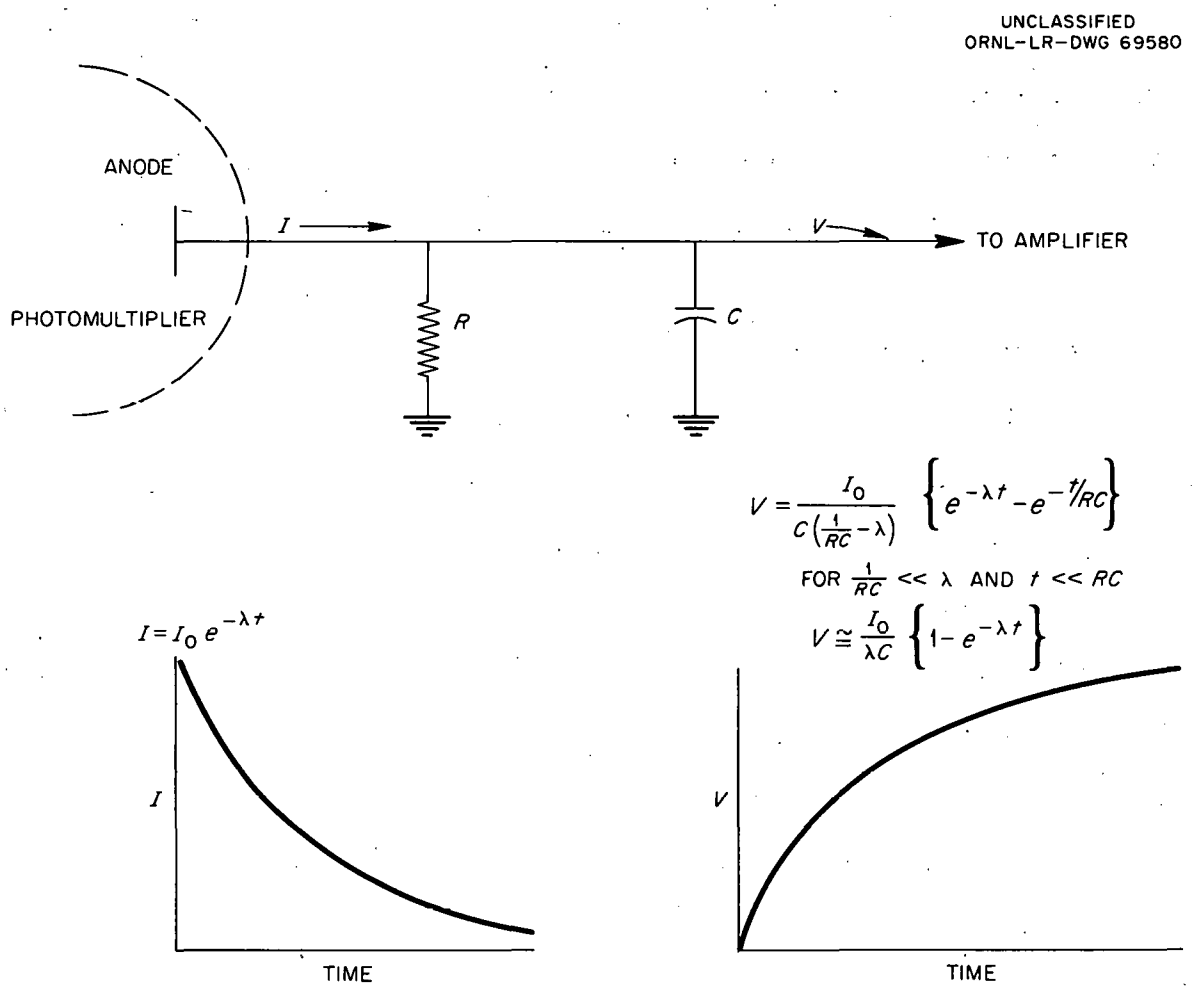

Fig. 6. The Anode Integrating Circuit Used in the Scintillation Experiments

was used with this system in order to insure that the pulse-gating signal arrived at the proper time. The pulse-height analyzer used with the A-8 amplifier was a Nuclear Data Model ND-120.

The pulse-height analyzer system was calibrated in the region of interest after each run using a precision pulse generator and a Rubicon laboratory potentiometer. The use of the pulse generator permits all pulse heights to be referred to a voltage standard. The results of the experiments then do not depend sensitively upon the long-term gain of the electronic system or a knowledge of the zero of the analyzer. The shape of the voltage pulse from the pulse generator was matched to the shape of the voltage pulse produced at the anode of the photomultiplier for excitation of the CsI(Tℓ) crystal by the 662-kev gamma ray. The output of the pulse generator was coupled to the preamplifier stage of the linear amplifier. The mercury relay of the pulse generator was operated at fifty-seven cycles per second. This eliminated the possibility that the voltage pulse measured was biased by being in phase with sixty-cycle noise in the electronic system.

IV. EXPERIMENTAL PROCEDURE

The fundamental scheme of the experiments consisted in the measurement of the light output of a CsI(Tℓ) crystal for various charged particles relative to the light output of the same crystal for a gamma-ray standard. The 662-kev gamma ray resulting from the decay of Cs^{137} was used as the standard. The light output of CsI(Tℓ) for excitation by the gamma-ray standard was measured in alternation with the light output

resulting from the other exciting radiations.

The results of a typical pulse-height measurement for the 662-kev gamma ray for a one-microsecond pulse-analysis time are shown in Figure 7. The relative number of counts in a channel is shown as a function of the channel number in which the count appeared; this is called the differential pulse-height distribution. The pulse height of the channel, corresponding to the midpoint of the full-energy peak at two-thirds the maximum value of the peak, was taken to be a measure of the light output of the CsI(Tℓ) crystal. The pulse height of a given channel corresponded to the output voltage of the pulse generator required for producing pulses in that channel. The differential pulse-height spectrum for the 10.7-kev gamma ray is shown in Figure 8 along with the calibration of the pulse-analysis system in the region of the full-energy peak of the gamma ray.

The gain of a photomultiplier tube has been shown to change as the average dynode current increases beyond a given value.[16] The average dynode current depends upon the gain of the photomultiplier and the rate at which photons are incident upon the photocathode. A series of experiments was performed to establish an upper limit for the average anode current which would be used in the scintillation experiments. The experiments consisted of measuring the ratio of the light output for two gamma rays. The count rate produced by a 59.7-kev gamma ray was kept constant during the experiments. The count rate produced by a 662-kev

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[16]L. Cathey, IRE Trans. on Nuclear Sci. NS-5, 3, 109 (1958).

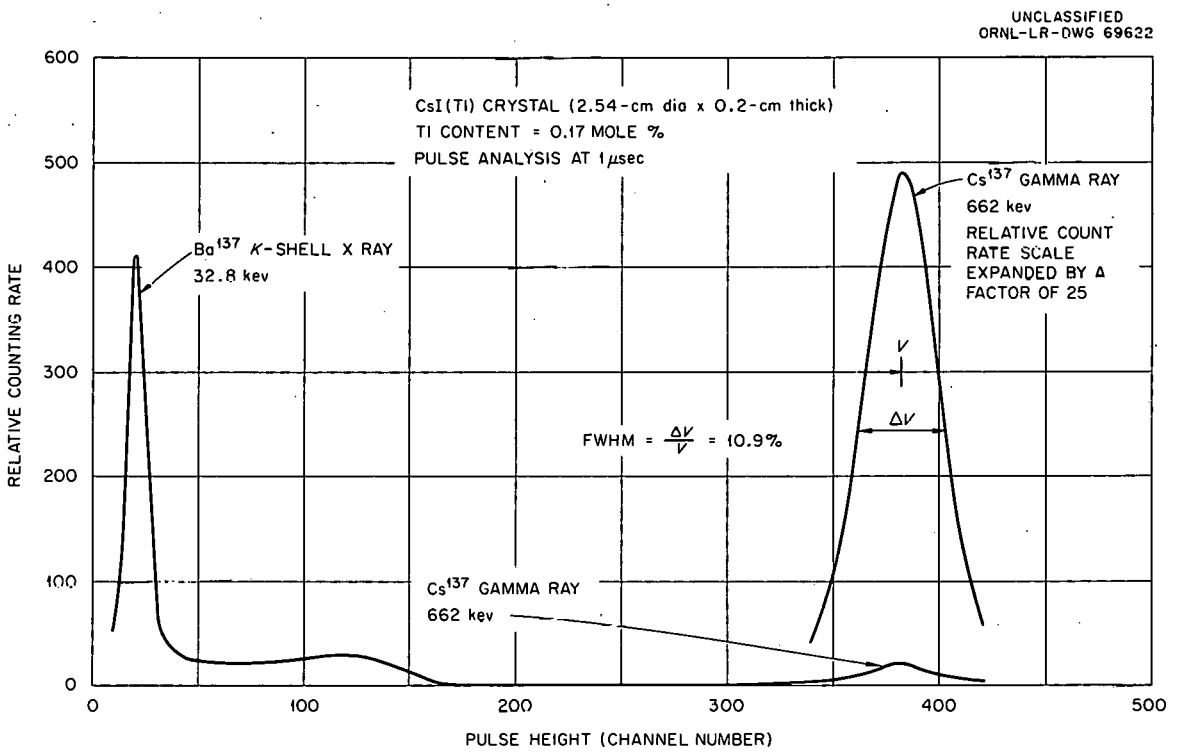

Fig. 7. Differential Pulse-Height Distribution Resulting from the Excitation of a CsI(Tℓ) Crystal with 662-kev Gamma Rays

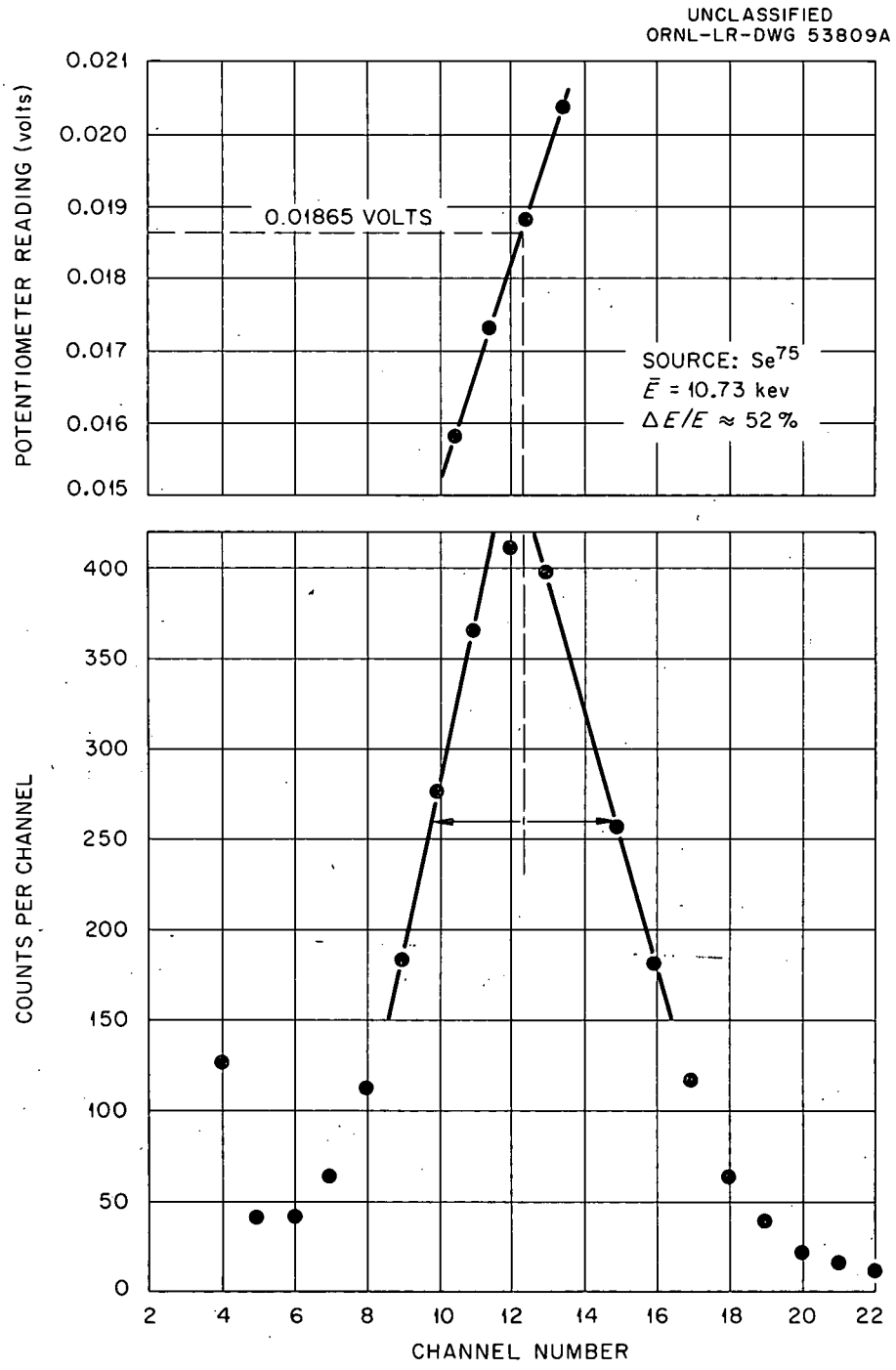

Fig. 8. The Differential Pulse-Height Distribution Resulting from the Excitation of a CsI(Tℓ) Crystal with 10.73-kev X rays, and the Calibration of the Pulse-Height Analyses

gamma ray was changed in successive experiments thereby producing a change in the average dynode current from one experiment to another. An interval of time elapsed between each experiment in order to allow the photomultiplier to recover from any effects due to a high anode current. For anode currents less than 0.1×10^{-6} amperes, the ratio of the light output of the two gamma rays was constant within the reproducibility of the measurements.

The pulse height corresponding to the full-energy peak was determined by plotting the counts in each channel as a function of channel number and fitting the best straight lines through the points on either side of the maximum (see Figure 8). The midpoint of these lines at two-thirds of the maximum of the distribution was taken to correspond closely with the mean pulse height in the full-energy peak. The midpoint of the two straight lines was chosen at two-thirds of the maximum because the second derivative of a Gaussian curve is zero at about two-thirds of its maximum value. The intersection of the straight lines through both sides of the peak was usually found to correspond to their midpoint as expected for a symmetric peak. In certain cases an asymmetric peak is expected. For example, in gamma-ray experiments the K-escape peak may cause the full-energy peak to be asymmetric. Corrections have been made for effects which are known to cause an asymmetric peak; these effects will be discussed later.

CHAPTER III

EXPERIMENTAL RESULTS

I. SCINTILLATION RESPONSE TO GAMMA RAYS

The scintillation response of the CsI(Tℓ) crystals was measured as a function of gamma-ray energy. The range of gamma-ray energies used extended from 10.7 kev to 2.7 Mev. The gamma-ray experiments were performed in order to obtain the scintillation response of CsI(Tℓ) to electrons. The gamma-ray experiments were considered to be more feasible than corresponding experiments using electrons to excite the crystal. Gamma rays interact with the atoms of the crystal and produce electrons in the conduction band. The interactions are the Compton scattering process, the photoelectric process, and pair production. The electrons so formed produce the ionization required for scintillation.

The crystals used in these experiments were too small (2.54 cm in diameter and 0.2 cm thick) to measure the full-energy peak accurately above 2.7 Mev. The full-energy peak becomes more difficult to measure for high-energy gamma rays because of the escape of Compton gamma rays from thin crystals. The fact that gamma rays having an energy greater than 2.7 Mev could not be measured in the present experimental program is not important, because the gamma-ray energy region of interest is below 1 Mev. The lower limit on the gamma-ray energy was fixed by the magnitude of the signal and the resulting width of the full-energy peak. Monoenergetic gamma rays resulting from nuclear transitions were used when

available. Below 100 kev some sources were used which emitted K-shell X rays as a result of internal conversion or by the K-capture process. The K-shell X rays are not monoenergetic since a K-shell vacancy can be filled by electrons from various levels in the higher shells (L, M, and N shells).

Although K-shell X rays are not monoenergetic, they can be used without introducing large uncertainties in the experimental results. Since the differential pulse-height distribution resulting from excitation of the CsI(Tℓ) crystal with K-shell X rays is a superposition of monoenergetic radiations, there is some uncertainty in associating an "average energy" for the source with the peak of the pulse-height distribution. The shape of the differential pulse-height distribution for K-shell X rays can be expressed as the sum of the Gaussian distributions resulting from each of the individual K-shell X ray components. The shape of the pulse-height distribution n(V) resulting from the superposition of Gaussian distributions is given by the relation

$$n(V) = \sum_i \frac{2 N_i \alpha_i}{\sqrt{\pi}} e^{-\alpha_i^2 (V-V_i)^2} . \qquad (4)$$

The relative intensity of the X rays associated with each Gaussian component is represented by N_i, and the term V_i represents the pulse height of a monoenergetic X ray of energy E_i. The term α_i is a measure of the line width of the pulse-height distribution resulting from X rays of energy E_i. The spread in energies of the K-shell X ray from a particular source is small; thus the variation of α_i is small and can be considered

constant. The pulse height V_p corresponding to the peak of the K-shell X ray differential pulse-height distribution can be obtained by equating the derivative of the pulse-height distribution given by Equation (2) to zero; this yields

$$V_p = \frac{\Sigma N_i V_i e^{-\alpha^2(V_p-V_i)^2}}{\Sigma N_i e^{-\alpha^2(V_p-V_i)^2}} . \qquad (5)$$

In order to define an average energy $\bar{E}$ characteristic of the K-shell X rays, the pulse height V_i was assumed to be given by $V_i = k E_i$ over the small energy range of the K-shell X rays from a given source. This is not a good assumption for K-shell X rays having energies near the K-absorption edge of the elements in the crystal (see Chapter IV). The equation for $\bar{E}$ is then

$$\bar{E} = \frac{\Sigma N_i E_i e^{-\alpha^2 k^2(\bar{E}-E_i)^2}}{\Sigma N_i e^{-\alpha^2 k^2(\bar{E}-E_i)^2}} . \qquad (6A)$$

For sufficiently small values of α, $\bar{E}$ approaches the "average energy" of the source E_s, which is given by the relation

$$E_s = \bar{E}_{\alpha \to 0} = \frac{\Sigma N_i E_i}{\Sigma N_i} . \qquad (6B)$$

Values of $\bar{E}$ were calculated from Equation (6A) using measured values of

k and α, and associated with the peak of the differential pulse-height distribution of K-shell X rays. The maximum that the energy defined by Equation (6A) differed from the energy given by Equation (6B) was 1 per cent.

The energies of the gamma rays used in the experiments are shown in Table I along with the radioactive isotope used to obtain the gamma rays. The average energy for K-shell X rays as calculated from Equation (6A) and Equation (6B) are also shown.

Values of $\overline{E}$ [Equation (6A)] are appropriate for the CsI(Tℓ) crystals having a thallium content of 0.17 and 0.046 mole per cent, and the values E_s [Equation (6B)] apply to the CsI(Tℓ) crystal having a thallium content of 0.002 mole per cent. The line width of the full-energy peak resulting from gamma-ray excitation of the crystal having a thallium concentration of 0.002 mole per cent was relatively large, and the values of α were thus small. For small values of α, Equation (6B) applies.

Two effects were considered which result in an asymmetric pulse-height distribution: (1) the escape of secondary X rays produced in the crystal by the primary gamma ray, and (2) the photomultiplier response for low values of the intensity of the incident light. For gamma-ray energies above the K-shell absorption energy, the escape of K-shell X rays results in another peak in the pulse-height distribution. This peak occurs at a pulse height which is less than the gamma-ray full-energy peak by an amount which is dependent upon the energy of the K-shell X ray. This escape peak may overlap the full-energy peak and produce an asymmetric peak. The measured full-energy peak was corrected, if necessary

TABLE I

GAMMA-RAY SOURCES

Radioactive Nuclide	Type of Radiation	$\bar{E}$ (kev)	E_s (a) (kev)	E_γ (b) (kev)
As^{75}	K-shell X ray	10.7	10.7	
Co^{57}	Nuclear gamma ray			14.37
In^{114}	K-shell X ray	24.7	24.7	
Xe^{131}	K-shell X ray	30.2	30.4	
Ba^{137}	K-shell X ray	32.7	32.88	
Yb^{170}	K-shell X ray	53.2	53.6	
Am^{241}	Nuclear gamma ray			59.72
Hg^{204}	K-shell X ray	71.75	72.5	
I^{131}	Nuclear gamma ray			80.2
Ce^{144}	Nuclear gamma ray			134
In^{114}	Nuclear gamma ray			191
Hg^{203}	Nuclear gamma ray			279
I^{131}	Nuclear gamma ray			364
Au^{198}	Nuclear gamma ray			411.8

TABLE I

(continued)

Radioactive Nuclide	Type of Radiation	$\bar{E}$ (kev)	E_s (a) (kev)	E_γ (b) (kev)
Cu^{164}	Annihilation radiation			511
Cs^{137}	Nuclear gamma ray			662
Zn^{165}	Nuclear gamma ray			1120
Na^{24}	Nuclear gamma ray			2750

a. The values of the relative intensities of the K-shell X ray components and the energy of these components were obtained from G. J. Nijgh, A. H. Wapstra, and R. Van Lieshout, Nuclear Spectroscopy Tables (North Holland Publishing Co., Amsterdam, 1959).

b. The values of the gamma-ray energies were obtained from D. Stominger, J. W. Holland, and G. T. Seaborg, Revs. Mod. Phys. 30, 585 (1958).

for the effect of the escape peak. The magnitude of the correction depends upon the energy of the gamma ray and the resolution of the system; the largest correction made for the effect of the escape peak was about 2.5 per cent. The correction was made by subtracting a Gaussian distribution characteristic of the escape peak from the full-energy peak. An estimate of the ratio of the counts in the escape peak to the counts in the full-energy peak was obtained from the work of Axel.[17] The pulse height obtained for gamma-ray excitation of the CsI(Tℓ) crystal having a thallium content of 0.002 mole per cent was relatively low, and the pulse-height distribution in the full-energy peak was asymmetric. The full-energy peak for very small pulse heights is not expected to be symmetric because of the statistical nature of both the interaction of photons with the photocathode to eject electrons and the ejection of secondary electrons at dynodes. For these cases, the maximum of the pulse-height distribution does not coincide with the mean value of the distribution; it is necessary to make a correction for this effect. A theoretical calculation of the pulse-height distribution at low pulse heights has been made by Wright.[18] The results of the calculation were expressed in terms of the mean number of electrons ejected from the photocathode by photons. The ratio F of the mean of the pulse-height distribution to the peak of the distribution was found to be

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[17]Peter Axel, Rev. Sci. Instr. 25, 391 (1954).

[18]G. T. Wright, J. Sci. Instr. 31, 462 (1954).

$$F = 1 + (\delta/N_m), \tag{7}$$

where N_m is the peak of the measured differential pulse-height distribution, and δ is a constant which depends upon the particular photomultiplier used. For a photomultiplier in which the dynode statistics for a single electron event were assumed to be characterized by a Poisson distribution, the value of δ was found to be 0.72.[18] Some experimental measurements were made which permitted the evaluation of δ by Prescott and Takhar.[19] The observed pulse distribution for a single electron incident on the first dynode was found to be nearly exponential. Values of δ obtained in the experiments ranged from 1.5 to 2.0 depending upon the mean number of electrons ejected from the photocathode.[20]

In order to make a correction for the asymmetric pulse-height distribution observed in the present work, it was necessary to measure the mean number of photoelectrons ejected in the scintillation events. This information was obtained from auxiliary experiments which consisted of measuring the electric current flowing to the photocathode for the steady-state excitation of the CsI(Tℓ) crystal by alpha particles. The only external connections to the photomultiplier were at the first dynode, the focussing electrode, and the photocathode. The first dynode was

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[19]J. R. Prescott and P. S. Takhar, IRE Trans. on Nuclear Sci. NS-9, 36 (1962).

[20]J. R. Prescott, University of Alberta, Canada, private communication.

held at a positive potential of 240 volts with respect to the photocathode, and a Keithley Electrometer model 610A was connected in series with the first dynode. The photomultiplier tube was mounted in a chamber which could be evacuated in order to use an alpha-particle source to produce the scintillations. With the focussing electrode adjusted to obtain the maximum current, the current to the photocathode was measured for a known counting rate produced by 4.8-Mev alpha particles from the decay of U^{233}. The current in the circuit was measured without the source present in order to make a correction for the background current. The mean number of electrons leaving the photocathode for a scintillation event produced by 4.8-Mev alpha particles was then obtained from a knowledge of the current to the photocathode and the scintillation counting rate. The mean number of electrons ejected from the photocathode per scintillation was found to be about 7×10^3 for alpha-particle (4.8 Mev) excitation of the CsI(Tℓ) crystal having a thallium content of 0.046 mole per cent. In order to estimate the mean number of electrons ejected from the photocathode for excitation of the crystal with 662-kev gamma rays, the number of photons produced in a scintillation was assumed to be proportional to the pulse height of the full-energy peak resulting from the radiation. The pulse height of the full-energy peak produced by the 4.8-Mev alpha particle was measured relative to the full-energy peak produced by the 662-kev gamma ray. The calculation of the mean number of electrons ejected from the photocathode for excitation of the crystal by gamma rays of other energies was assumed to depend upon a linear relation of the pulse height as a function of energy.

The mean number of electrons ejected from the photocathode for the lowest pulse height observed was estimated to be about nineteen. The value of δ used for the correction of the data in the present experiments was 1.3 ± 0.6 which represents the average of the values quoted by Prescott[20] and Wright.[18] The largest correction made amounted to 6.8 per cent of the pulse height of the full-energy peak in the case of the Ba^{137} K-shell X ray exciting the crystal having a thallium content of 0.002 mole per cent.

The experimental data from the gamma-ray experiments are shown in Figures 9 and 10, where the relative pulse height per unit energy is shown as a function of the gamma-ray energy. The curves are normalized to unity at 662 kev. Figures 9 and 10 show the data obtained for pulse-analysis times of one and seven microseconds respectively. The error limits are shown for a few points on the curves and include uncertainties due to the measurements, the corrections to the full-energy peaks, and knowledge of the gamma-ray energy. The same type of behavior shown in Figures 9 and 10 for CsI(Tℓ) has been observed for NaI(Tℓ).[21-25] The shapes of the curves shown in Figures 8 and 9 do not depend appreciably upon

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[21]D. Engelkemeir, Rev. Sci. Instr. 27, 589 (1956).

[22]W. W. Managan, Paper 5.10 of Proc. Sixth Tripartite Instr. Conf., AECL-805 (1959).

[23]P. Iredale, Nucl. Instr. and Methods 11, 340 (1961).

[24]J. A. Nemilov, J. J. Lomonosov, A. N. Pesoreveski, L. V. Soshin, and E. D. Teterin, Izvestia Academia Nauk, SSSR, 24, 257 (1959).

[25]T. H. Jones, Nucl. Instr. and Methods 15, 55 (1962).

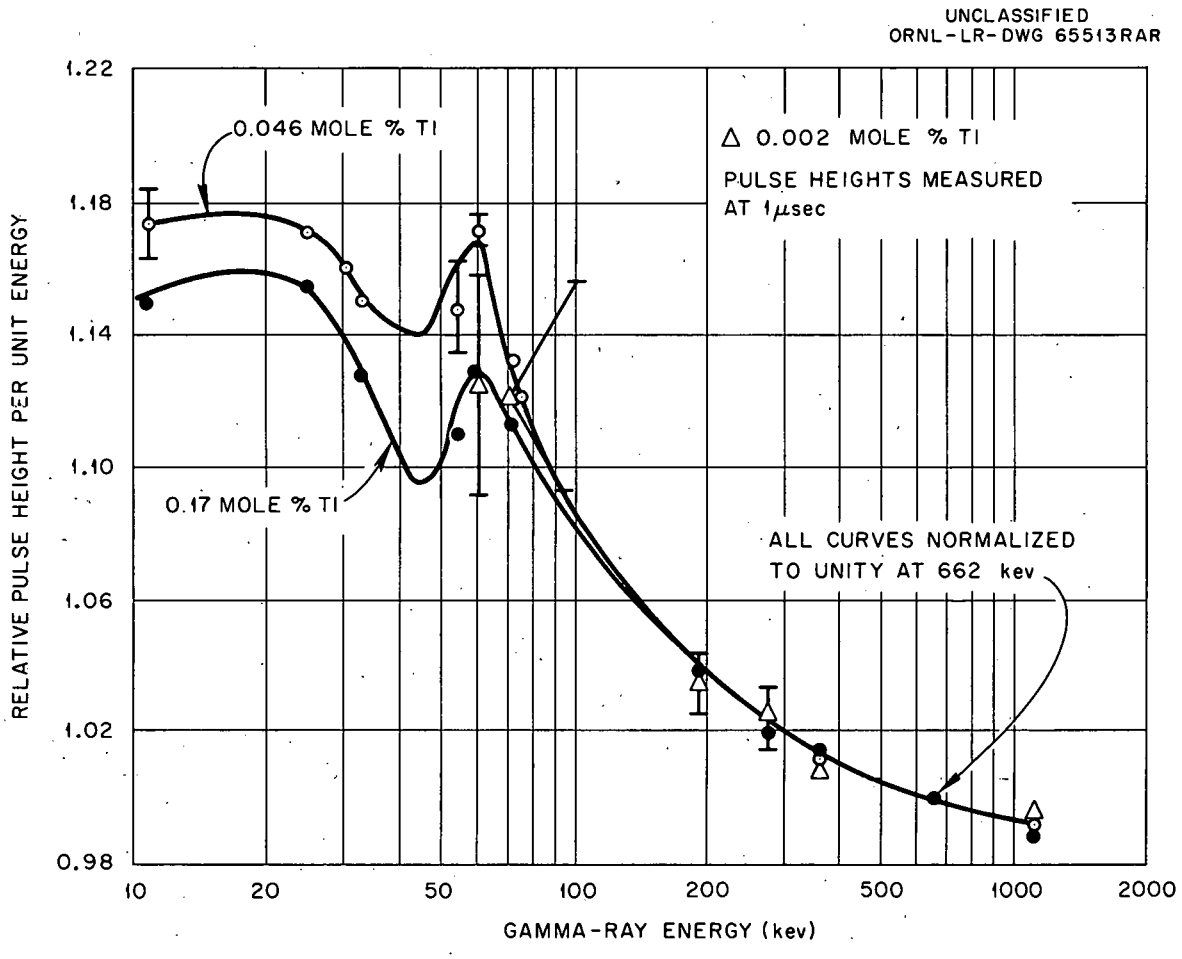

Fig. 9. Measured Pulse Height Per Unit Energy for Gamm Rays on CsI(Tℓ) Crystals of Varying Thallium Content for a Pulse-Analysis Time of 1 μsec

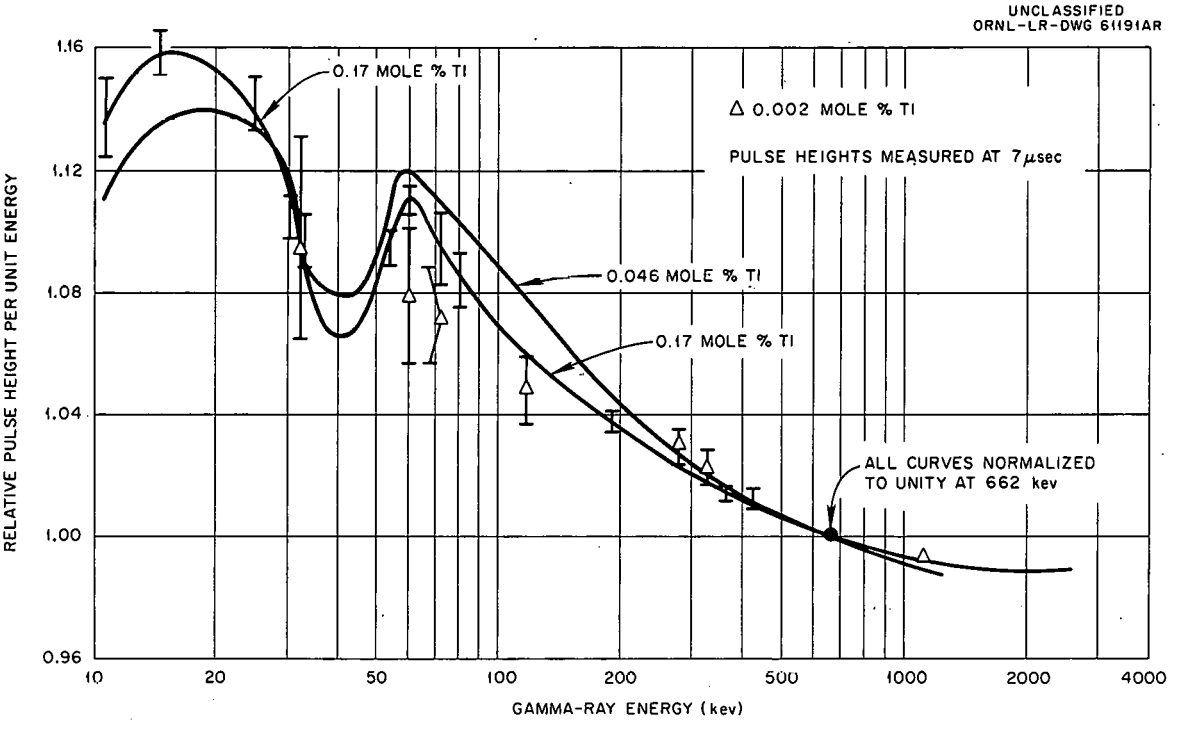

Fig. 10. Measured Pulse Height Per Unit Energy for Gamma Rays on CsI(Tℓ) Crystals of Varying Thallium Content for a Pulse-Analysis Time of 7 μsec

the thallium concentration within the accuracy of the experiments. This is in agreement with the results of the scintillation model.

The relative pulse height per unit energy is greater at low gamma-ray energies for the one-microsecond pulse-analysis time than for the seven-microsecond pulse-analysis time. This result is a consequence of the fact that the time dependence of the scintillation pulse depends upon the ionization density produced by the exciting particle.[9]

The light output of CsI(Tℓ) crystals having various thallium concentrations was measured for excitation by 662-kev gamma rays in order to obtain the ratio of the macroscopic absorption cross section $N_a\sigma_a$ for thallium-luminescence centers to the macroscopic absorption cross section $N_\ell\sigma_\ell$ of lattice traps, since this ratio appears in the scintillation model. A theoretical expression for the relative luminescence efficiency as a function of activator concentration has been given by Johnson and Williams.[6] This expression was used in the scintillation model in the form

$$L \sim \frac{C(1 - C)^z}{C + \frac{\sigma_\ell}{\sigma_a}(1 - C)} \qquad (8)$$

where C is the mole fraction of the activator, and z is an effective number of lattice sites surrounding a given activator such that concentration quenching will occur if another activator atom is contained within z.

The data obtained from the measurement of the light output as a function of the thallium content are shown in Figures 11 and 12 for pulse-analysis times of 1.25 and seven microseconds respectively. These data were used in Equation (8) to determine values of σ_a/σ_ℓ and z. The

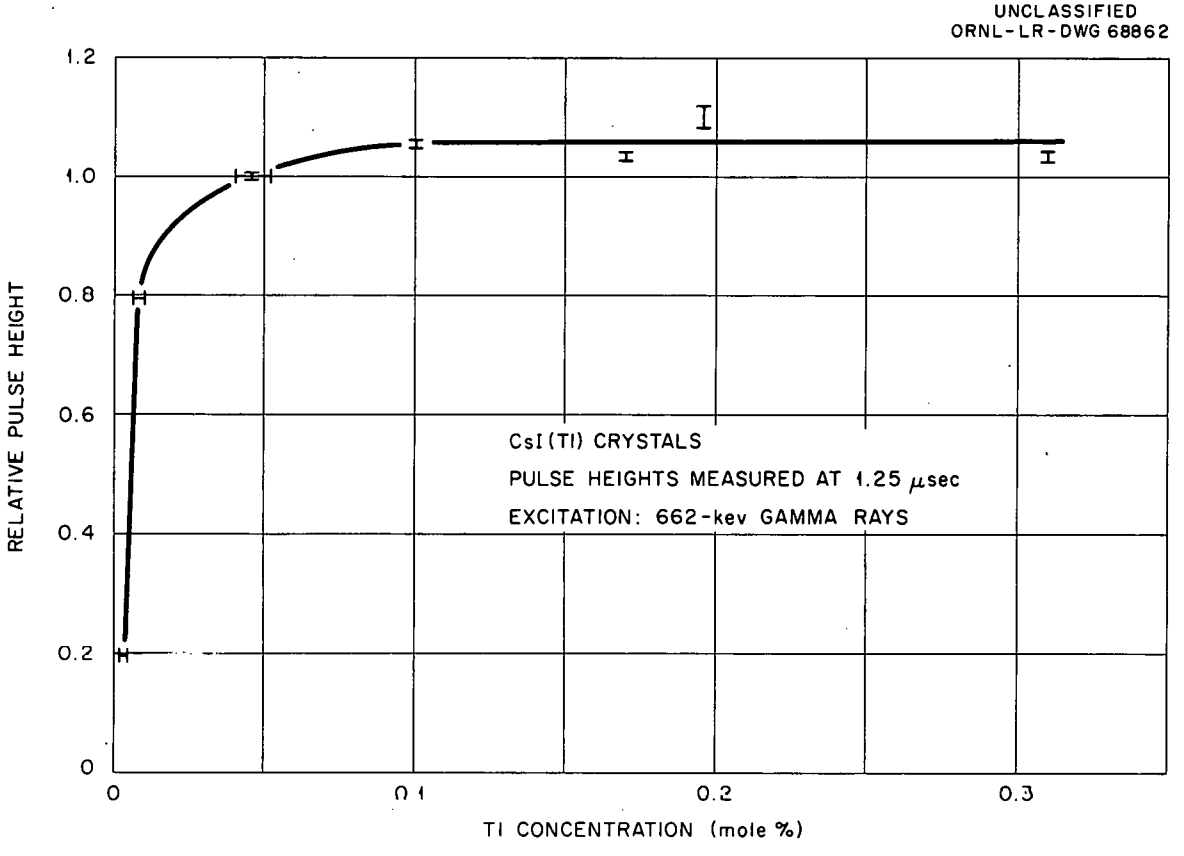

Fig. 11. Relative Pulse Height Resulting from the Excitation of CsI(Tℓ) Crystals with 662-kev Gamma Rays as a Function of the Thallium Content of the Crystal. The voltage pulse was measured at 1.25 μsec

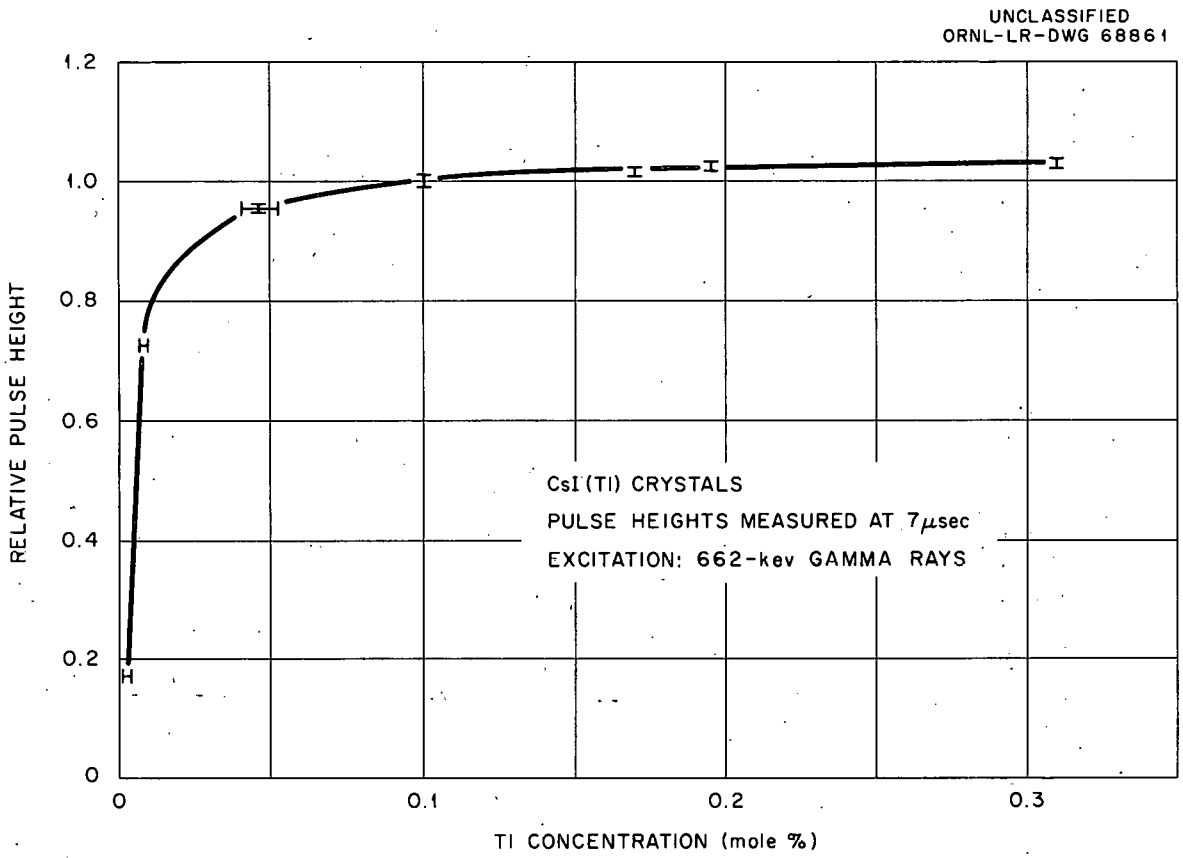

Fig. 12. Relative Pulse Height Resulting from the Excitation of CsI(Tℓ) Crystals with 662-kev Gamma Rays as a Function of the Thallium Content of the Crystal. The voltage pulse was measured at 7 μsec

values found for σ_a/σ_ℓ and z were 0.22×10^{-3} and 65, respectively, for the data obtained from a pulse-analysis time of 1.25 microseconds; for a pulse-analysis time of seven microseconds the corresponding values were 0.16×10^{-3} and 44.

II. SCINTILLATION RESPONSE TO PROTONS

The scintillation response of four CsI(Tℓ) crystals to monoenergetic protons was measured over the energy range from 0.2 to 5.5 Mev. Monoenergetic protons were obtained with the Oak Ridge National Laboratory 5.5-MV Van de Graaff generator.

The primary beam from the Van de Graaff generator was deflected by a magnetic field through an angle of 90 degrees into a scattering chamber. The protons struck a thick Ta^{181} target, and a portion of the scattered beam entered the entrance slit of the 60-degree charged-particle analyzing magnet. The energy of the protons which pass through the exit slit and impinge upon the CsI(Tℓ) crystal was determined by the magnetic field of the analyzing magnet.

Both the 90-degree bending magnet and the 60-degree charged-particle analyzing magnet were calibrated prior to performing the scintillation response experiments. The energy of charged particles which are deflected through a given angle by the magnets was determined by the relation

$$E = K H^2, \tag{9}$$

where H is the magnetic field, and K is a constant. The values of K for

a particular magnet have been found to be independent of the magnetic field over the range of magnetic fields used in the present experiments.[26] The 90-degree bending magnet was calibrated with protons whose energy corresponded to the threshold of the $Li^7(p,n)$ reaction; the threshold of this reaction occurs at 1880.7 kev.[27] The 60-degree charged-particle analyzing magnet was calibrated by scattering protons having the energy of the $Li^7(p,n)$ threshold from the Ta^{181} target through an angle of 90 degrees into the analyzing magnet. The calibrations of the two magnets were then checked by an intercomparison procedure in order to provide assurance that the values of K were independent of the magnetic field. In this procedure the energy of protons which were analyzed by the 60-degree charged-particle analyzing magnet was determined from the measured magnetic field of the 90-degree bending magnet and the angle through which the protons were scattered to enter the analyzing magnet. The energy of the protons being analyzed by the 60-degree magnet and the value of the magnetic field deflecting these protons were used to determine values of K [Equation (9)] for the 60-degree magnet. The values of K for the 60-degree charged-particle bending magnet were constant within 0.2 per cent for a range of energies from 1.0 to 3.5 Mev. This confirmed the fact that the values of K are essentially independent of the magnetic field.

The charged particle leaving the exit slits of the analyzing magnet passed through the 0.32-cm hole in the hemispherical shell of the

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[26]J. K. Bair, personal communication.

[27]J. Marion, Revs. Modern Phys. 33, 139 (1961).

photomultiplier assembly and struck the CsI(Tℓ) crystal being investigated. The distribution of scintillation events in the crystals for the charged-particle experiments was not the same as for the 662-kev gamma-ray experiments. The 662-kev gamma rays excite the crystals over their entire volume while the charged particles excite only a volume of the crystal near its surface and confined to the 0.32-cm diameter circle at the center of the top surface. An experiment was performed to investigate the possibility that there was a detectable difference in the optical collection efficiency for the two cases. Gamma rays from Am^{241} having an energy of 59.72 kev were collimated and introduced into the photomultiplier assembly used for charged particles and struck the same area of the crystal that the charged particles did. The pulse height of the 59.72-kev gamma ray was measured relative to that of the 662-kev gamma-ray standard. The relative pulse height of the 59.72-kev gamma ray was the same as measured in the regular assembly used for gamma rays. More than one-half of the 59.72-kev gamma rays were absorbed in the first 0.02 cm of the crystal and any significant variations in the optical collection efficiency for scintillations produced by the gamma-ray standard and the charged particles would have been observed in this experiment.

The experimental results on the scintillation response of CsI(Tℓ) crystals to protons are shown in Figures 13, 14, and 15 for the crystals having thallium contents of 0.002, 0.046, and 0.17 mole per cent, respectively. The pulse height per unit energy is shown as a function of the proton energy. Each figure shows the results obtained for both pulse-analysis times. The pulse height per unit energy for protons for

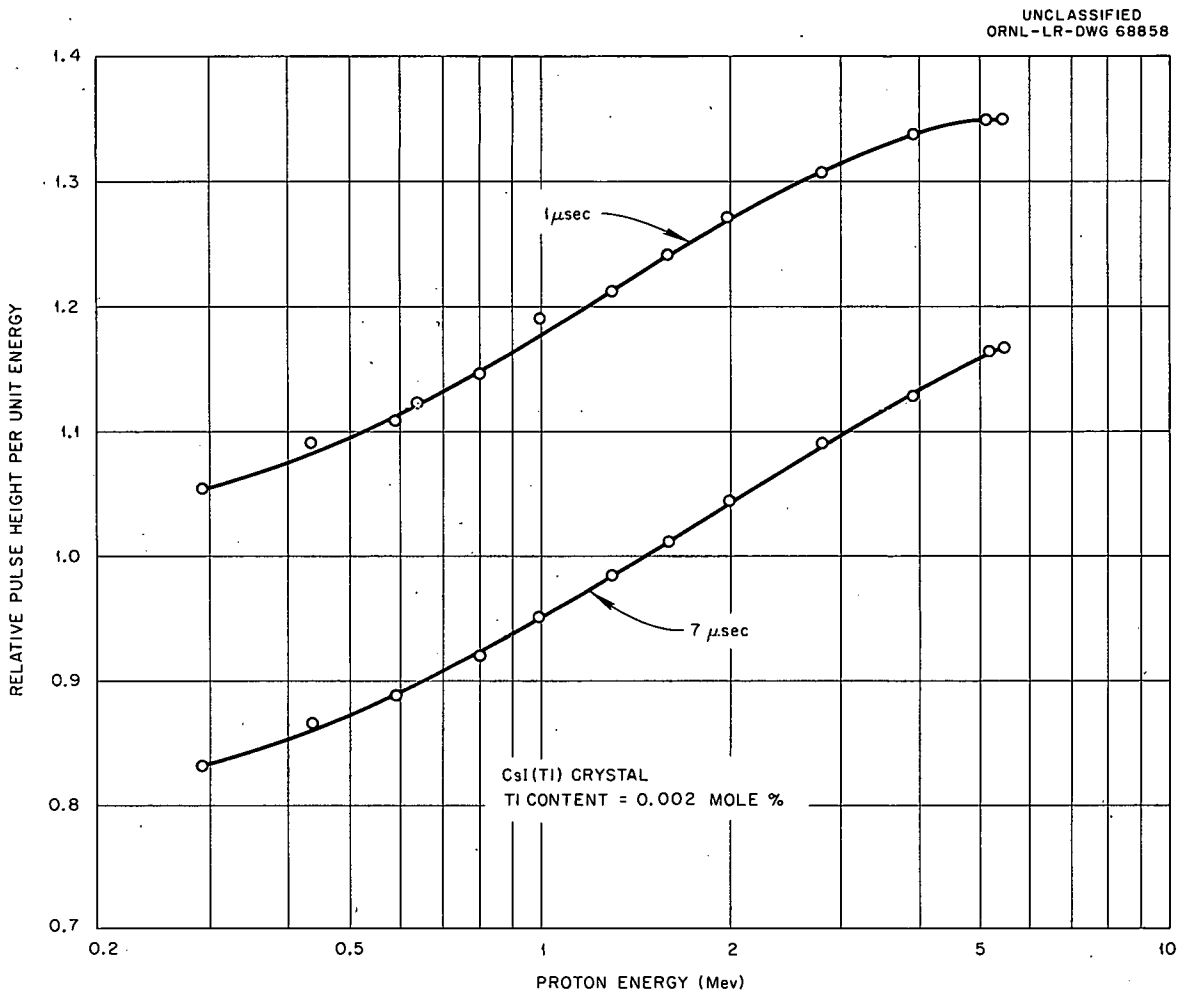

Fig. 13. Measured Values of Pulse Height Per Unit Energy, $(L/E)_p$, for Proton Excitation of a CsI(Tl) Crystal Having a Thallium Content of 0.002 Mole Per Cent. All values of $(L/E)_p$ are normalized to $(L/E)_\gamma = 1$, for E_γ = 662 kev

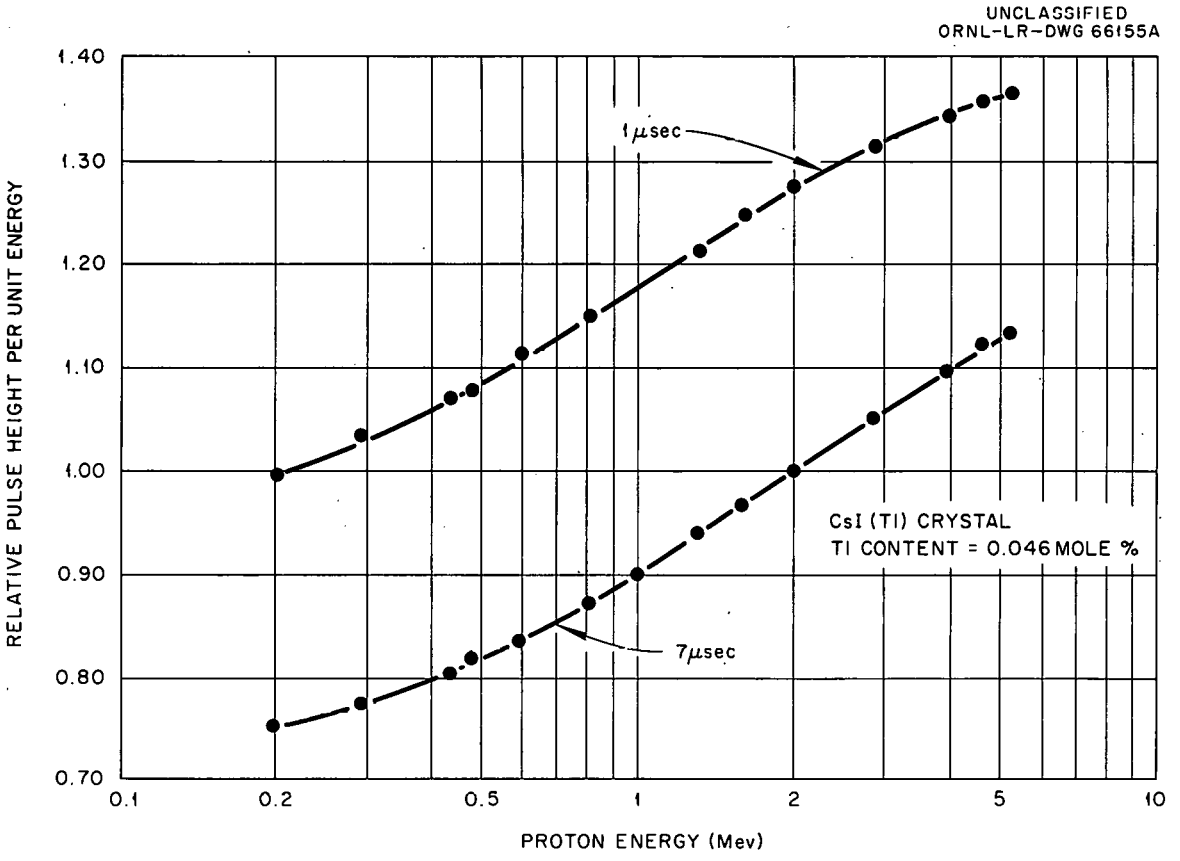

Fig. 14. Measured Values of Pulse Height Per Unit Energy, $(L/E)_p$, for Proton Excitation of a CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent. All values of $(L/E)_p$ are normalized to $(L/E)_\gamma = 1$, for E_γ = 662 kev

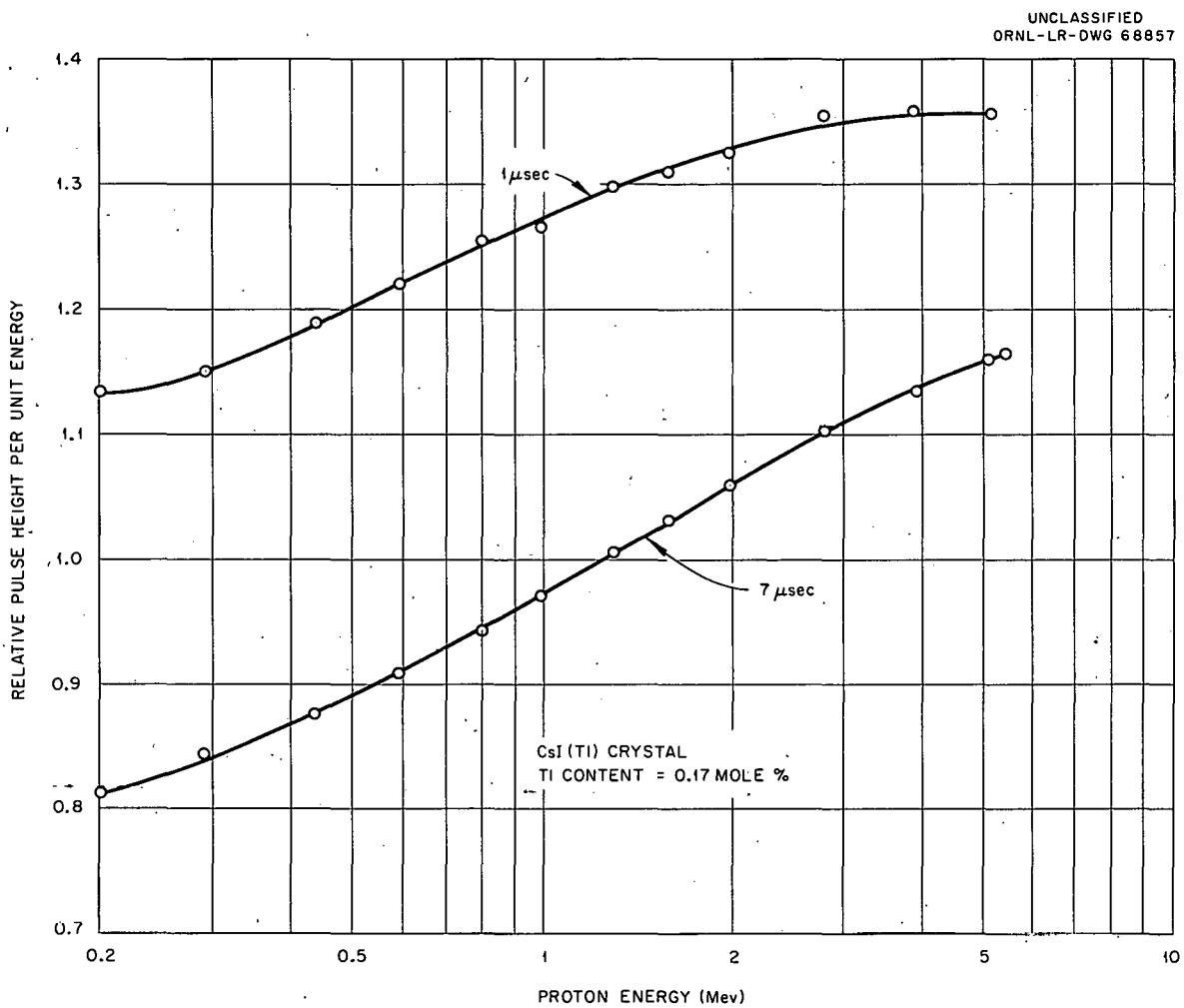

Fig. 15. Measured Values of Pulse Height Per Unit Energy, $(L/E)_p$, for Proton Excitation of a CsI(Tℓ) Crystal Having a Thallium Content of 0.17 Mole Per Cent. All Values of $(L/E)_p$ are normalized to $(L/E)_\gamma = 1$, for E_γ = 662 kev

a given pulse-analysis time is normalized relative to the pulse height per unit energy at the same pulse-analysis time for the 662-kev gamma rays. The experimental data show that for a pulse-analysis time of one microsecond, the pulse height per unit energy for protons at 662 kev is greater than that for gamma rays at 662 kev; for a pulse-analysis time of seven microseconds the converse is true. During the course of the experiments, the amplified pulses produced by 662-kev gamma rays and 662-kev protons were observed simultaneously, and the proton pulse was observed to have a faster initial rise than the gamma-ray pulse. At one microsecond the proton pulse was greater than the gamma-ray pulse, while at seven microseconds the gamma-ray pulse was greater than the proton pulse. This is in accordance with detailed measurements of the time dependence of scintillation pulses from CsI(Tℓ) excited by particles producing different ionization densities.[9] These observations demonstrate that the detailed nature of the relative scintillation response of CsI(Tℓ) to charged particles is a complicated function of the pulse-analysis time and energy of the radiations.

An examination of the data in Figures 13, 14, and 15 clearly shows that the light output of CsI(Tℓ) to proton excitation is not proportional to the energy of the protons over the entire energy range investigated. A proportional response of light output versus energy would be a horizontal line for the representation of pulse height per unit energy versus energy. A detailed examination of the data shows that light output as a function of energy is also nonlinear over most of the energy range investigated.

III. SCINTILLATION RESPONSE TO ALPHA PARTICLES

The scintillation response to alpha particles was measured for the crystals having thallium contents of 0.046 and 0.31 mole per cent. The energy of the alpha particles ranged from 58 kev to 10 Mev. The Oak Ridge National Laboratory 5.5-MV Van de Graaff generator was used in conjunction with the 60-degree charged-particle analyzing magnet in the same manner as for the proton experiments. Alpha-particle energies above 5 Mev were obtained by accelerating doubly-charged helium ions. The experimental procedure was the same as for the other cases; that is, the pulse height corresponding to an alpha particle of energy E_α was measured relative to the pulse height of the 662-kev gamma ray from Cs^{137}.

Since the shape of the scintillation pulse from CsI(Tℓ) is dependent upon the ionization density of the exciting radiation, an investigation was made to determine the effect of changing the shape of the pulser pulse on the measured scintillation response of alpha particles with respect to the scintillation response of gamma rays. The experiment consisted of measuring the light output of a CsI(Tℓ) crystal for excitation by alpha particles and the 662-kev gamma rays at the two pulse-analysis times for different shapes of the pulser pulse. In one case the shape of the pulser pulse was made to resemble the gamma-ray pulse in the seven-microsecond region. In another case the shape of the pulser pulse was made to resemble the alpha-particle pulse over the first two microseconds of the pulse. No change in the relative values of the alpha-particle pulse heights to those of the gamma rays for the two pulse-analysis times was observed. Within the accuracy of the experiments, it

was concluded that the differences in pulse shapes presented to the analyzing system in these experiments did not affect the results of the experiments, as expected.

The experimental results of the measurements on the scintillation response of the CsI(Tℓ) crystal having a thallium content of 0.046 mole per cent to alpha particles are shown in Figure 16, where the pulse height per unit energy is shown as a function of the energy of the alpha particles. The data from both pulse-analysis times are shown. The minimum in the curves of Figure 16 occurs in the energy region for alpha particles where the stopping power of CsI for these particles is at a maximum. According to the scintillation model, a minimum in the pulse height per unit energy versus energy curve will occur for excitation by any charged particle as it passes through the energy region where a maximum in the stopping power occurs. This feature results because the model treats the scintillation efficiency dL/dE as a continuous and monotonic function of the stopping power dE/dx at large values of dE/dx. The results of the experiments on alpha particles are also shown in Figure 17, where the pulse height L is shown as a function of the alpha-particle energy; the data are shown on two different scales. The kink in the pulse height versus energy curve corresponds to the minimum of the pulse height per unit energy versus energy curve. The light output of CsI(Tℓ) crystals is clearly not a linear function of energy for alpha particles.

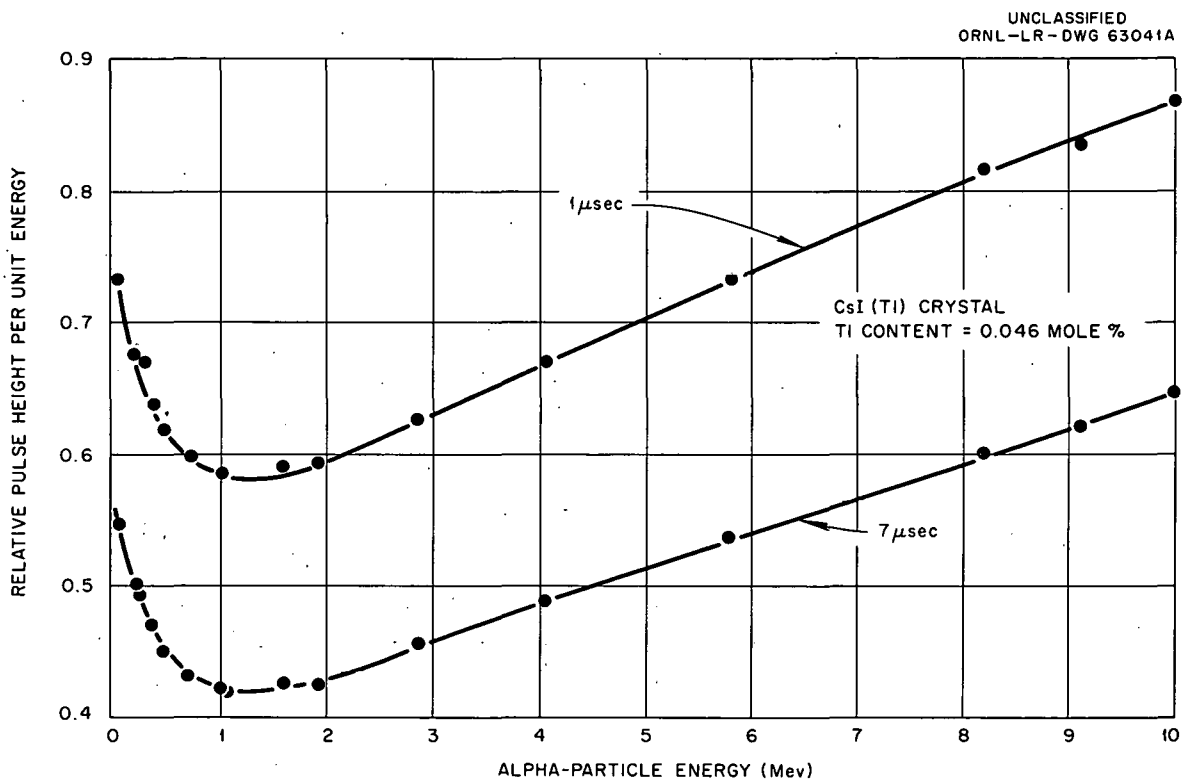

Fig. 16. Measured Values of Pulse Height Per Unit Energy, $(L/E)_\alpha$, for Alpha-Particle Excitation of the CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent. The curves are normalized to $(L/E)_\gamma = 1$, for $E_\gamma = 662$ kev

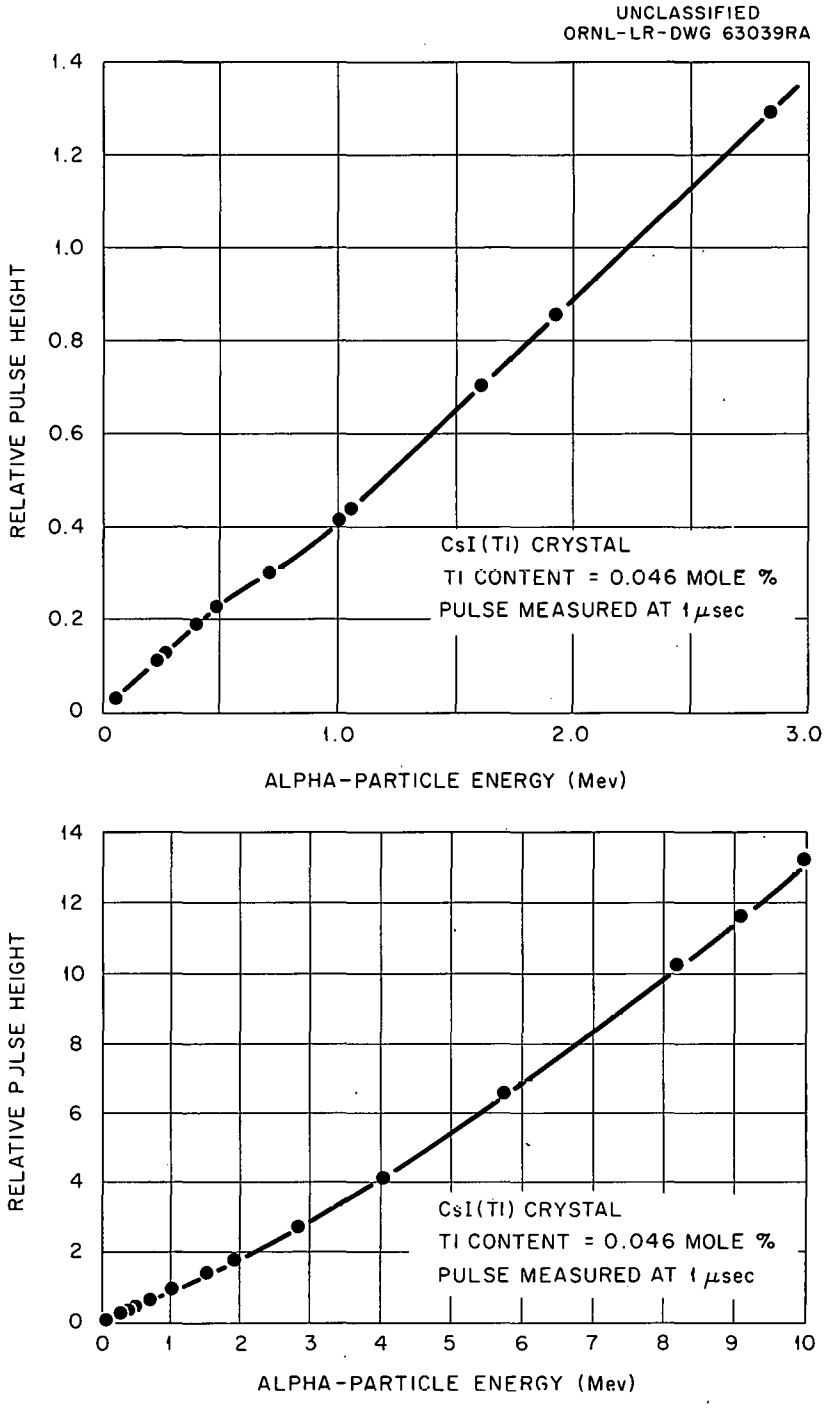

Fig. 17. The Relative Pulse Height for Alpha-Particle Excitation of the CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent. The scales of the ordinates are different in the two cases

IV. EMISSION SPECTRUM OF CsI(Tℓ)

The emission spectra of CsI(Tℓ) crystals were measured for several thallium concentrations and for various exciting radiations. The thallium content of the crystals ranged from zero (no thallium intentionally added to the crystals) to 0.17 mole per cent. The exciting radiations were 50- and 250-kev X rays, 1.4- and 4.4-Mev protons, and 2.0- and 8.7-Mev alpha particles. Hrehuss measured the emission spectra of a CsI(Tℓ) crystal under excitation by electrons, deuterons, and alpha particles and observed that the emission spectrum depended upon the exciting radiation.[13] Differences in the emission spectra must be considered in the analysis of the data obtained in the measurement of the scintillation response of CsI(Tℓ) crystals for various charged particles.

The emission spectra were measured with a Bausch and Lomb 500-millimeter focal-length grating monochromator which had a nominal dispersion of 33 angstroms per millimeter. The grating was blazed for the ultraviolet of the first order. The light transmitted by the monochromator was detected by an RCA-7265 photomultiplier tube. This photomultiplier tube has a multi-alkali photocathode with an S-20 response. The current output of the photomultiplier tube was measured with a Keithley model 610 electrometer; the output signal of the electrometer was continually registered on a chart recorder. A synchronous motor turned the wavelength control drum of the monochromator at a scanning speed of about fifteen A per second. The relative spectral response of the monochromator-photomultiplier system was measured using a lamp, having a tungsten-ribbon

filament, which was calibrated by the National Bureau of Standards. The X ray machine used in the measurements of the emission spectra was supplied with filters to provide a reasonably limited band of X ray energies.[28] The 250-kev X ray band extended from about 150 kev to 275 kev and the 50-kev band extended from about 30 kev to 50 kev.

The charged particles for use in the measurements of the emission spectra were obtained with the 5.5-MV Van de Graaff generator at the Oak Ridge National Laboratory. In the experiments using the charged particles to excite the CsI(Tℓ) crystals it was necessary to monitor the total light emitted by the crystal because of fluctuations in the intensity of the beam of charged particles. An RCA-6342 photomultiplier tube was used to detect the total light, and the current output of the tube was measured by a Keithley model 610R electrometer. The output signal of the electrometer was registered continually by a chart recorder. The current density on the surface of the crystals was about 10^{-9} amperes per cm^2.

The results of the measurements of the emission spectra are shown in Figures 18, 19, 20, and 21 where the relative intensity is shown as a function of the wavelength. Figures 18, 19, and 20 show the emission spectra of the CsI(Tℓ) crystals having thallium contents of 0.002, 0.046, and 0.17 mole per cent, respectively, for excitation by various radiations. Figure 21 shows the emission spectra for CsI(Tℓ) crystals of different thallium contents for excitation by 4.4-Mev protons. The spectral distribution curves are normalized to the same number of photons.

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[28]J. C. Villforth, R. D. Birkhoff, and H. H. Hubbell, Jr., "Comparison of Theoretical and Experimental Filtered X Ray Spectra," ORNL-2529, 46 (1958).

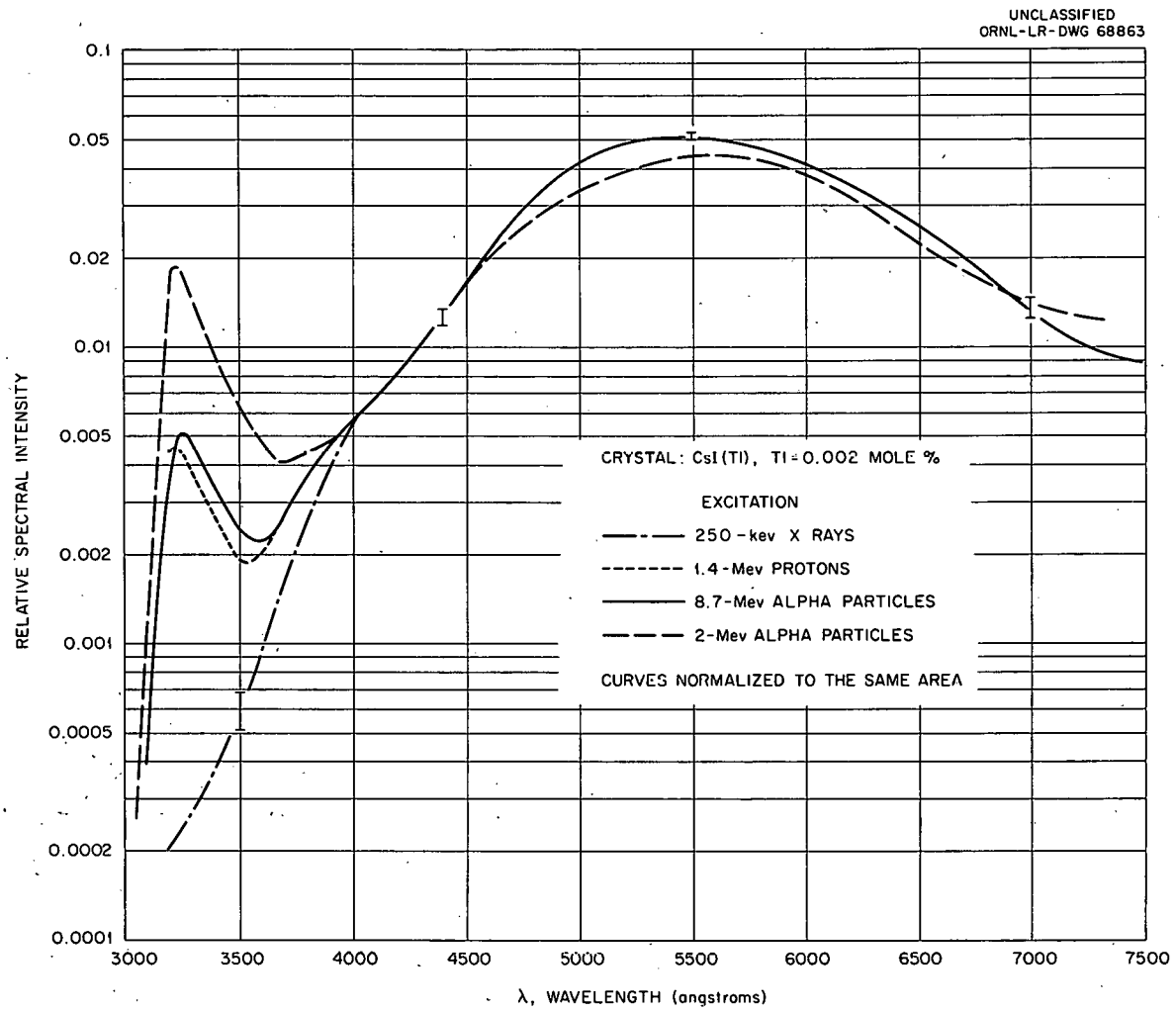

Fig. 18. The Emission Spectra of a CsI(Tℓ) Crystal Having a Thallium Content of 0.002 Mole Per Cent for Excitation by Various Radiations. The curves for the 250-kev X rays, 1.4-Mev protons, and 8.7-Mev alpha particles merge at about 4000 angstroms

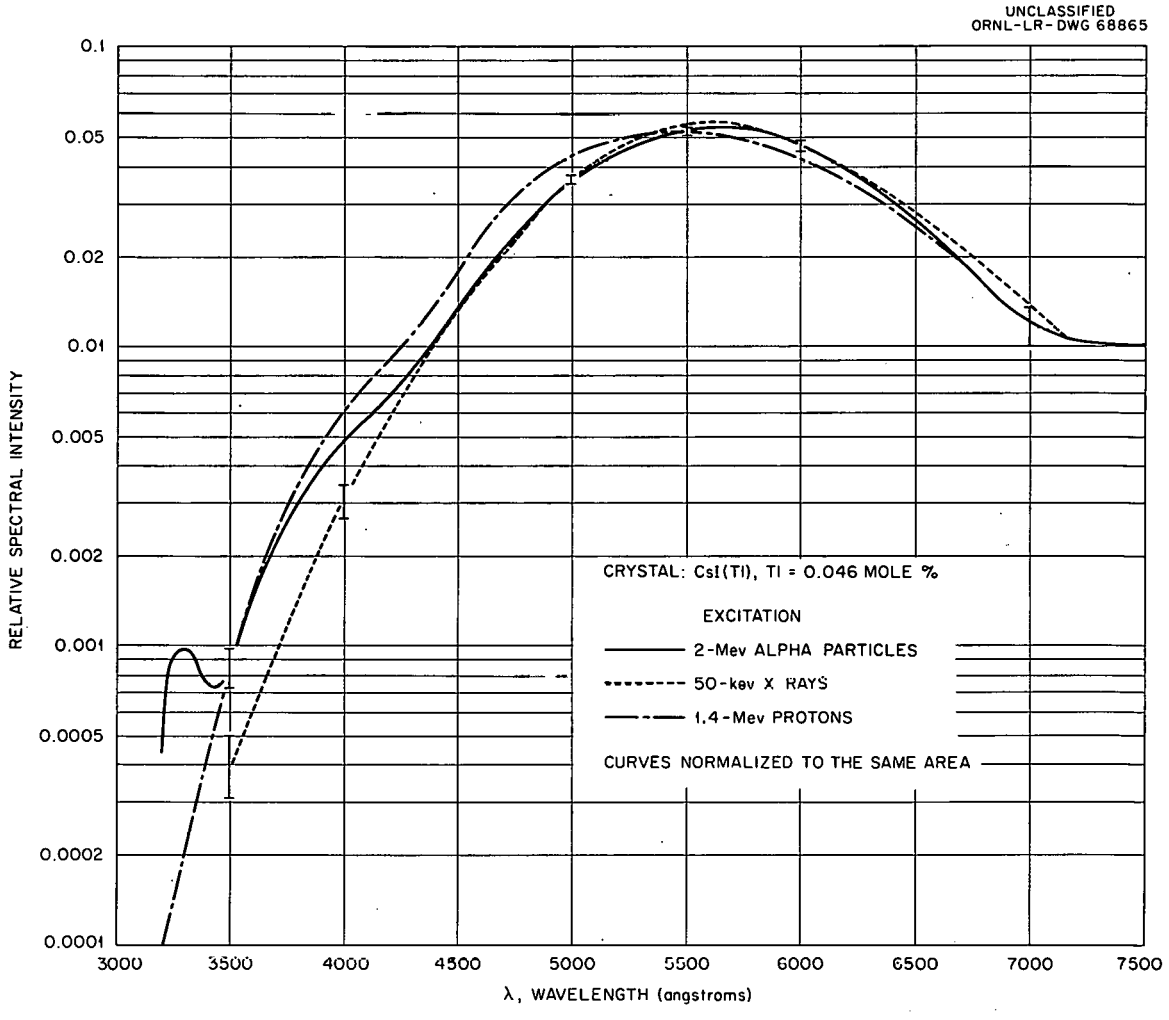

Fig. 19. The Emission Spectra of a CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent for Excitation by Various Radiations

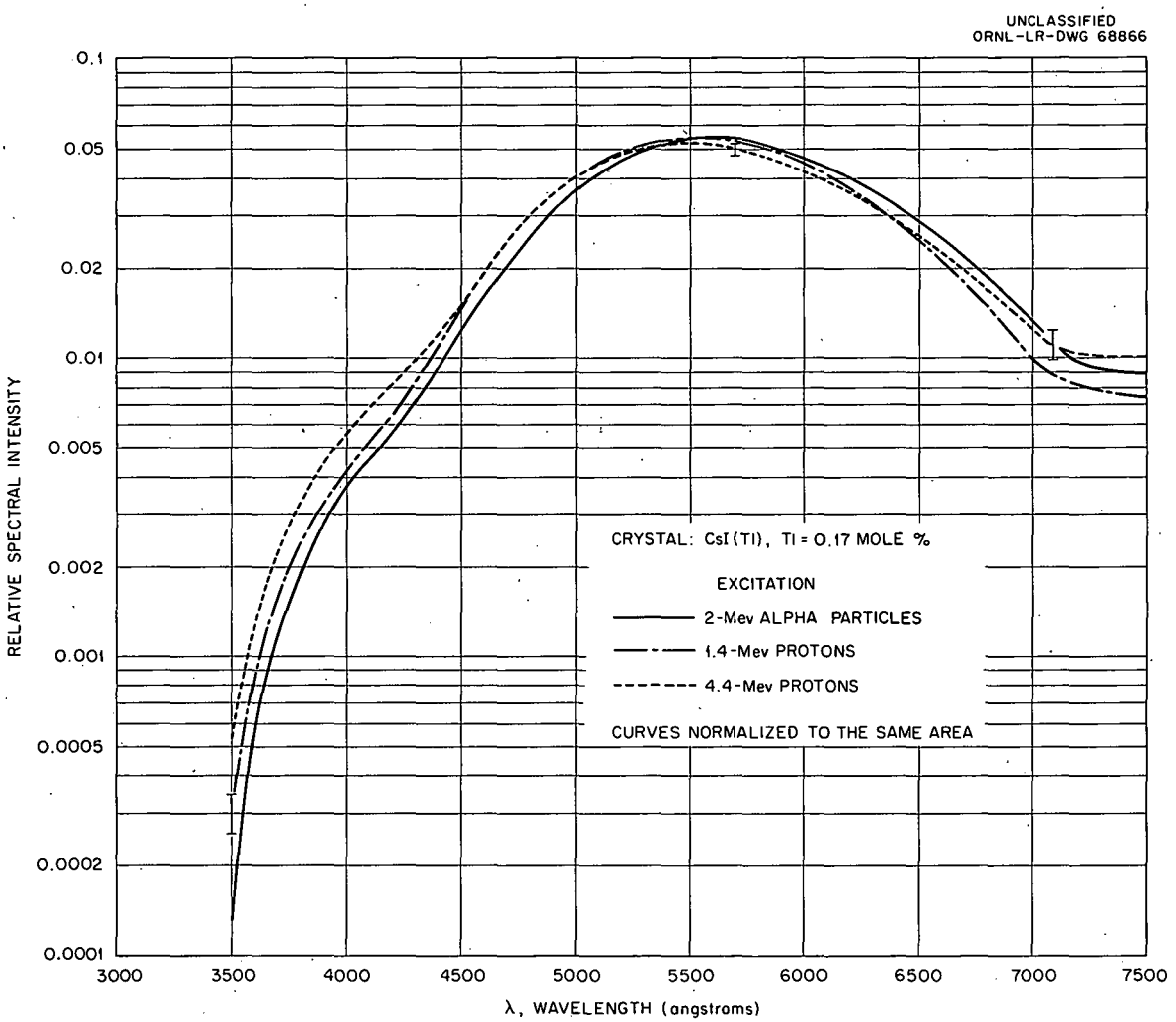

Fig. 20. The Emission Spectra of a CsI(Tℓ) Crystal Having a Thallium Content of 0.17 Mole Per Cent for Excitation by Various Radiations

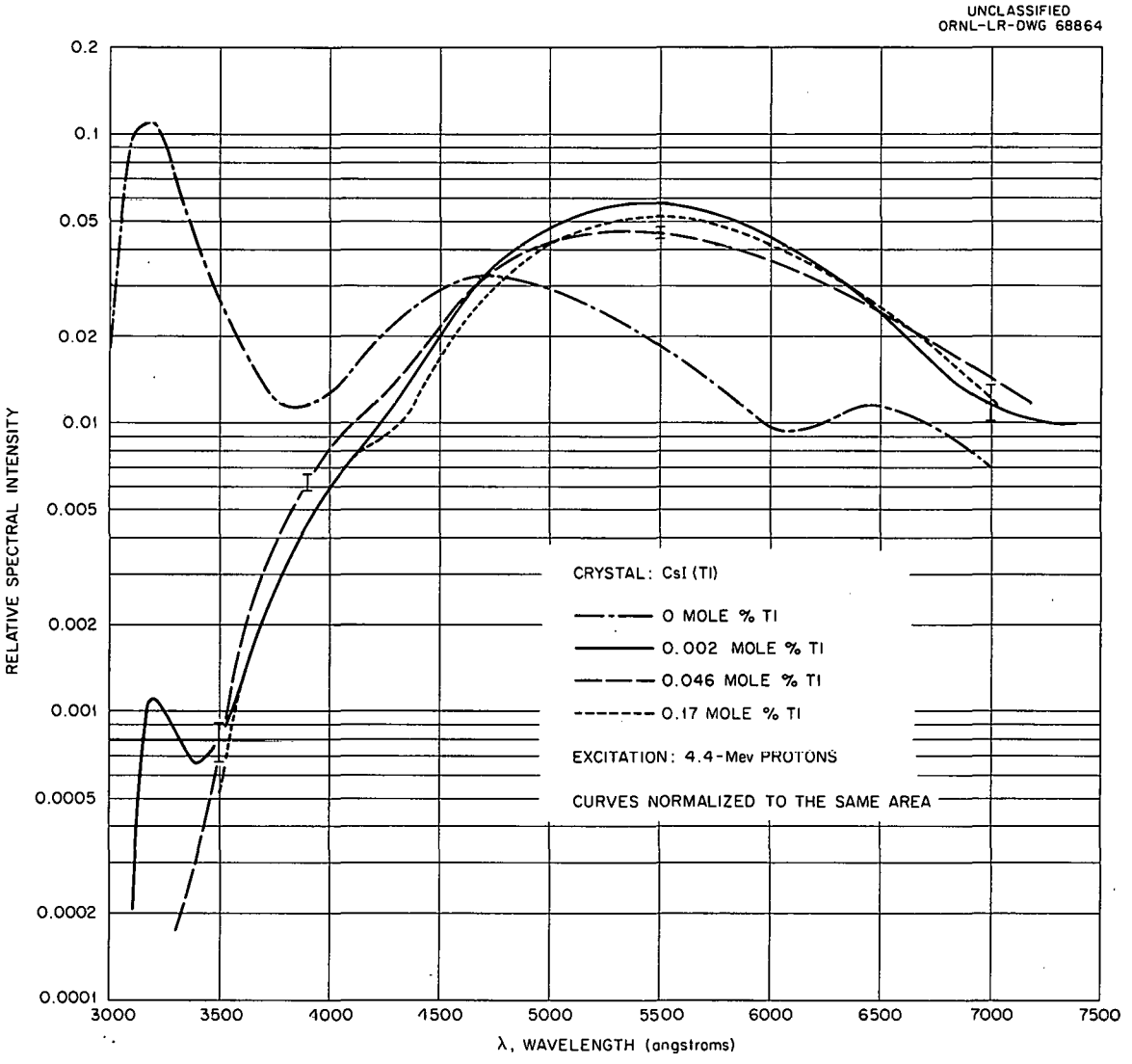

Fig. 21. The Emission Spectra of CsI(Tℓ) Crystals Having Various Thallium Contents for Excitation by 4.4-Mev Protons

Spectral measurements above 7500A were limited by the transmission of the monochromator and the sensitivity of the photomultiplier tube. The lower limit of about 3100A was set by the absorption of the light by the glass of the photomultiplier tube. The broad emission band of CsI(Tℓ) observed in the present work has been reported by other investigators.[4,13] The spectral region measured in the present work covers a wider range of wavelengths than has been reported previously. The emission spectra measured in this work have been corrected for the spectral sensitivity of the detector system.

The current output of a photomultiplier tube resulting from a scintillation pulse is proportional to the integral over the emission spectrum of the product of the relative emission intensity and the spectral sensitivity of the photomultiplier tube. The relative response S for a given photomultiplier assembly is given by the relation

$$S = K \int I(\lambda)\ P(\lambda)\ d\lambda \qquad (10)$$

where $I(\lambda)$ is the spectral intensity at a wavelength λ; $P(\lambda)$ is the relative sensitivity of the photomultiplier tube for light of wavelength λ; and K is a proportionality constant. The quantity $P(\lambda)$ was assumed to be the spectral sensitivity of the photomultiplier tube for light incident upon the face of the tube. Values of S/K were calculated from Equation (10) for the emission spectra measured in the present work, using an S-20 type response curve for $P(\lambda)$. The shape of the S-20 response (see Figure 3) was taken from the specifications given by the maker (Radio

Corporation of America) of the tube type used in the experiments. The relative values of S/K were found to be constant within the uncertainty of the measurements. The maximum variation in S/K was about 4 per cent with most of the values falling within a range of about 2 per cent. Since the values of S/K were found to be nearly constant for the nominal S-20 response, the spectral sensitivity of the photomultiplier used in the scintillation experiments was not measured. The 3300A band is not considered to be due to the thallium activator; in order to obtain precise comparison of the experiments with the scintillation model, the contribution of the 3300A band was subtracted from the measured scintillation response. Although values of S/K for this latter case differed from the previous ones, the values fell within a range of about 3 per cent. This means that the pulse heights observed in the scintillation response experiments can be used as a direct measure of the light output characteristic of the thallium luminescence which is at a maximum at about 5700A. This result is due to the broad spectral response of the photomultiplier tube used and the fact that most of the light emitted by CsI(Tℓ) is above 5000A.

The fact that an emission band appears at 3300A for CsI(Tℓ) crystals at room temperature complicates the interpretation of the scintillation experiments on CsI(Tℓ). Experiments performed by various investigators have shown that the luminescence band characteristic of CsI(Tℓ) crystals is excited by ultraviolet light in the region of 3200A.[4,12,29]

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[29]R. G. Lagu and B. V. Thosar, Proc. Indian Acad. Sci. 53A, 219 (1961).

Forro[30] measured the absorption peaks of CsI(Tℓ) due to the presence of thallium and observed an absorption peak at 2990A; the long wavelength tail of the absorption peak extended above 3300A. The experiments of Knoepfel, Loepfe, and Stoll[4] showed that the excitation spectrum from thallium-activator luminescence extended to 3500A for certain CsI(Tℓ) crystals but extended only to about 3350A for other crystals. It must be concluded that some thallium-activator luminescence produced by charged particles is excited in CsI(Tℓ) crystals by photons emitted by a luminescence center which is not associated with thallium. This same observation has been made with respect to NaI(Tℓ) by Van Sciver.[31]

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[30]M. Forro, Z. Physik 58, 613 (1929).

[31]W. J. Van Sciver, Phys. Rev. 120, 1193 (1960).

CHAPTER IV

DERIVATION OF THE SCINTILLATION RESPONSE FOR ELECTRONS

The scintillation response of CsI(Tℓ) to electrons can be calculated from the measured gamma-ray response using a model outlined by Zerby, Meyer, and Murray.[32] The light output of the crystal due to gamma-ray excitation originates from the ionization produced in the crystal by electrons which result from gamma-ray interactions in the crystal.

The model of Zerby, Meyer, and Murray[32] was based on gamma-ray interactions in a sodium iodide crystal. In the gamma-ray energy region of interest only electrons in iodine ions were assumed to participate in photoelectric events; the photoelectron cross sections are much greater for iodine than for sodium. Three energy levels were used, corresponding to the K-shell, L-shell, and M-shell energy levels. In a photoelectric interaction a gamma ray was assumed to interact with an electron in the deepest energy level for which the interaction was energetically possible. In a photoelectric event gamma rays having energies E_γ which are greater than the K-shell binding energy E_K eject an electron with a kinetic energy $E_e = E_\gamma - E_K$ from the K-shell. The K-shell vacancy was assumed to be filled by an L-shell electron resulting in a K-shell X ray which in turned ejected an L-shell electron; this leaves two vacancies in the

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[32]C. D. Zerby, A. Meyer, and R. B. Murray, Nuclear Instr. and Methods 12, 115 (1961).

L-shell. An L-shell vacancy was assumed to be filled by processes which yield an average light output B. The number of photons L_γ emitted by the crystal due to a photoelectric event with a K-shell electron can be written in the form

$$L_\gamma = C_e[E_\gamma - E_K] + A + 2B \tag{11}$$

where C_e is the light output per unit energy for an electron having an energy $E_e = E_\gamma - E_K$, and both A and B are constants. The constant A represents the light output resulting from the process of filling the K-shell vacancy produced by the incident gamma ray. The quantity B represents the light output resulting from the filling of an L-shell vacancy and the subsequent processes. In a similar manner the light output from photoelectric events due to gamma rays having energies above the L-shell binding energy but below the K-shell binding energy can be expressed in the form

$$L_\gamma = C_e(E_\gamma - E_L) + B \tag{12}$$

where E_L is the L-shell binding energy. It may be noted that for an iodine K-shell X ray source $L_\gamma = A + B$.

In the present work the model of Zerby, Meyer, and Murray can be used provided the interactions of gamma rays with both cesium and iodine are considered. The K-shell and L-shell absorption energy levels of iodine and cesium are comparable. The K-shell binding energy of cesium

is about 36 kev and that of iodine is about 33.2 kev. About 54 per cent of the K-shell photoelectric interactions occur in cesium, and 46 per cent occur in iodine. The presence of thallium was not considered because of its low concentration in the crystal. Since the K-shell binding energies in cesium and iodine are reasonably close together, a single K-shell absorption level for CsI(Tℓ) crystals was used; the energy of this level was chosen to be the average of the cesium and iodine levels weighted with their relative K-shell photoelectric cross sections. This is a good approximation because the energies of the electrons ejected from the cesium or iodine K-shell are only about 2.8 kev apart, and $(L/E)_e$ is a slowly varying function of the electron energy. The K-shell absorption energy used in the calculations was 34.7 kev. The same procedure was used for the L-shell binding energy, and a value of 4.2 kev was used for the L-shell absorption energy.

For gamma-ray energies below 80 kev only photoelectric events were assumed to occur. At 80 kev about 5 per cent of the gamma-ray interactions are other than photoelectric events, and at 60 kev essentially all of the interactions are photoelectric events. For gamma-ray energies of about 80 kev or less, Equations (11) and (12) were used with the appropriate values of E_K and E_L to find the scintillation response of CsI(Tℓ) crystals for excitation by electrons. The solution of Equations (11) and (12) was obtained by successive iterations in which a curve of the pulse height per unit energy [the quantity C_e in Equations (11) and (12)] for electrons was assumed, and the light output of the crystal for gamma rays was calculated using Equations (11) and (12). The iterative

procedure was simplified by the knowledge of the scintillation response of CsI(Tℓ) to K-shell X rays characteristic of cesium and iodine. The quantity A + B represents the amount of light produced in the crystal by a mixture of cesium and iodine K-shell X rays external to the CsI(Tℓ) crystal. For cesium or iodine K-shell X rays external to the crystal, the L-shell X ray emitted in the process was absorbed by the aluminum over the crystal before reaching the crystal. The quantity A + B can then be written as

$$A + B = 0.54\ L_{Cs} + 0.46\ L_{I} \tag{13}$$

where L_{Cs} and L_{I} represent the light output produced by cesium and iodine K-shell X rays external to the crystal. Values of L_{Cs} and L_{I} were obtained from the experiments performed in this investigation.

Since the normalized pulse height per unit energy for gamma rays or electrons is not greatly different from unity an estimate of the value of B can be obtained. The quantity B represents the light output produced in the process of filling a vacancy in the L-shell of an atom in the crystal, and a total energy of 4.2 kev is expended in the crystal in the filling of the L-shell vacancy and in the subsequent process. A first approximation to B was obtained by assuming that the L-shell vacancy was filled by a process in which a single 4.2-kev electron resulted and that the value of C_e for this electron was unity; thus B = 4.2. Using this value for B and the value derived from the experiments for A + B, values of C_e were calculated directly from Equations (11) and (12). The value

of B was then adjusted in order to obtain values of C_e which are consistent with both Equations (11) and (12).

For gamma-ray energies above 100 kev the calculational procedure was performed on a digital computer using a Monte Carlo technique.[32] The electron response curve which was derived for gamma-ray interactions below about 80 kev was extended to 1000 kev using the scintillation response of NaI(Tℓ) to electrons as a guide.[32] This curve was then used in the Monte Carlo procedure to calculate the scintillation response of the crystal for gamma rays having energies greater than 100 kev, and these values were compared with the results of the experiments of the present work. The electron response curve above 80 kev was modified on the basis of this comparison, and another Monte Carlo calculation was made. This procedure continued until the calculated gamma-ray response was the same as the experimentally measured response within the uncertainty of the gamma-ray experiments. The derived curves of the pulse height per unit energy for electrons as a function of the electron energy are shown in Figures 22 and 23 for pulse-analysis times of one and seven microseconds, respectively.

The uncertainty limits shown in Figures 22 and 23 are based on experimental uncertainties in the gamma-ray measurements and on estimates of the uncertainties introduced by the model used for the gamma-ray interactions in the crystal. Although the energy at which the pulse height per unit energy reaches a maximum is not well defined, a good estimate of the maximum value of dL/dE can be obtained from Figures 22

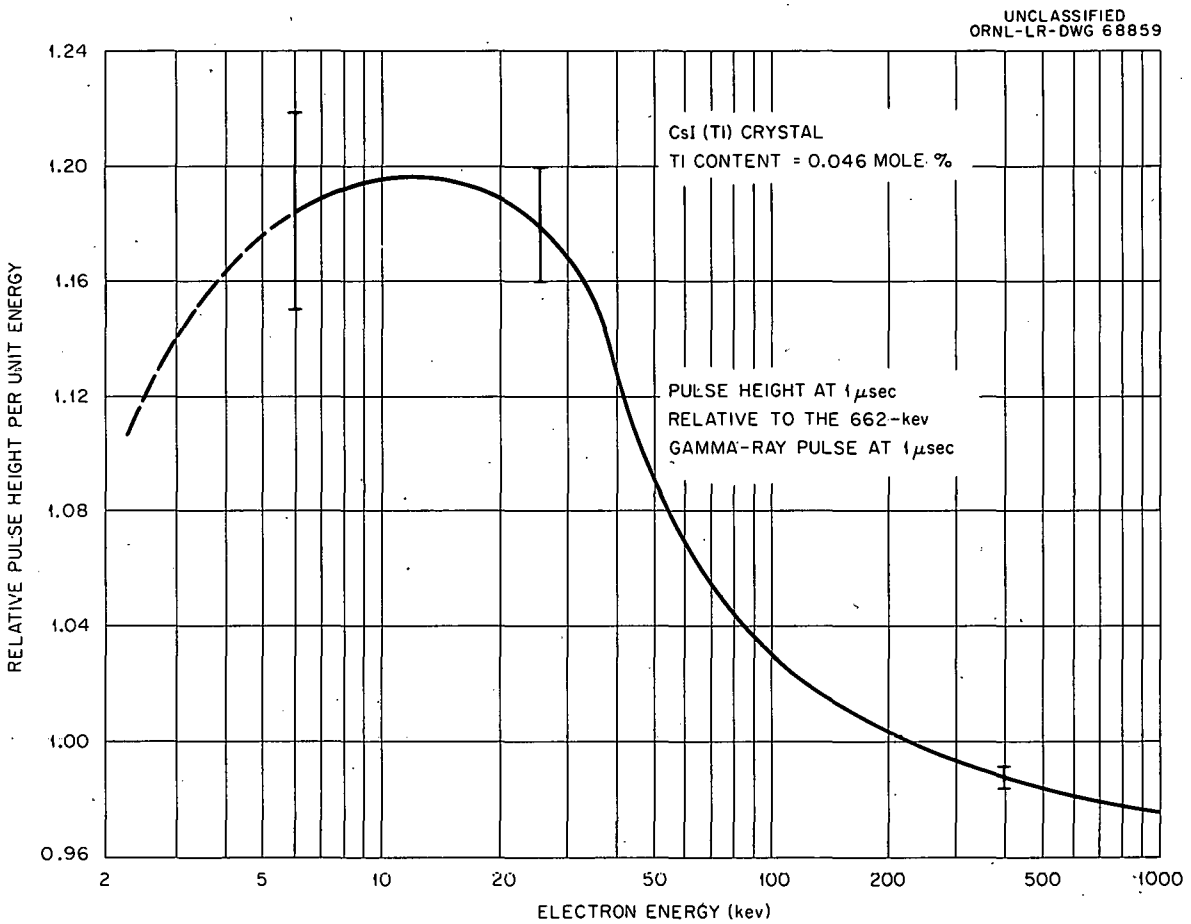

Fig. 22. Derived Value of the Pulse Height Per Unit Energy for Electron Excitation of a CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent. The dashed curve indicates large uncertainties.

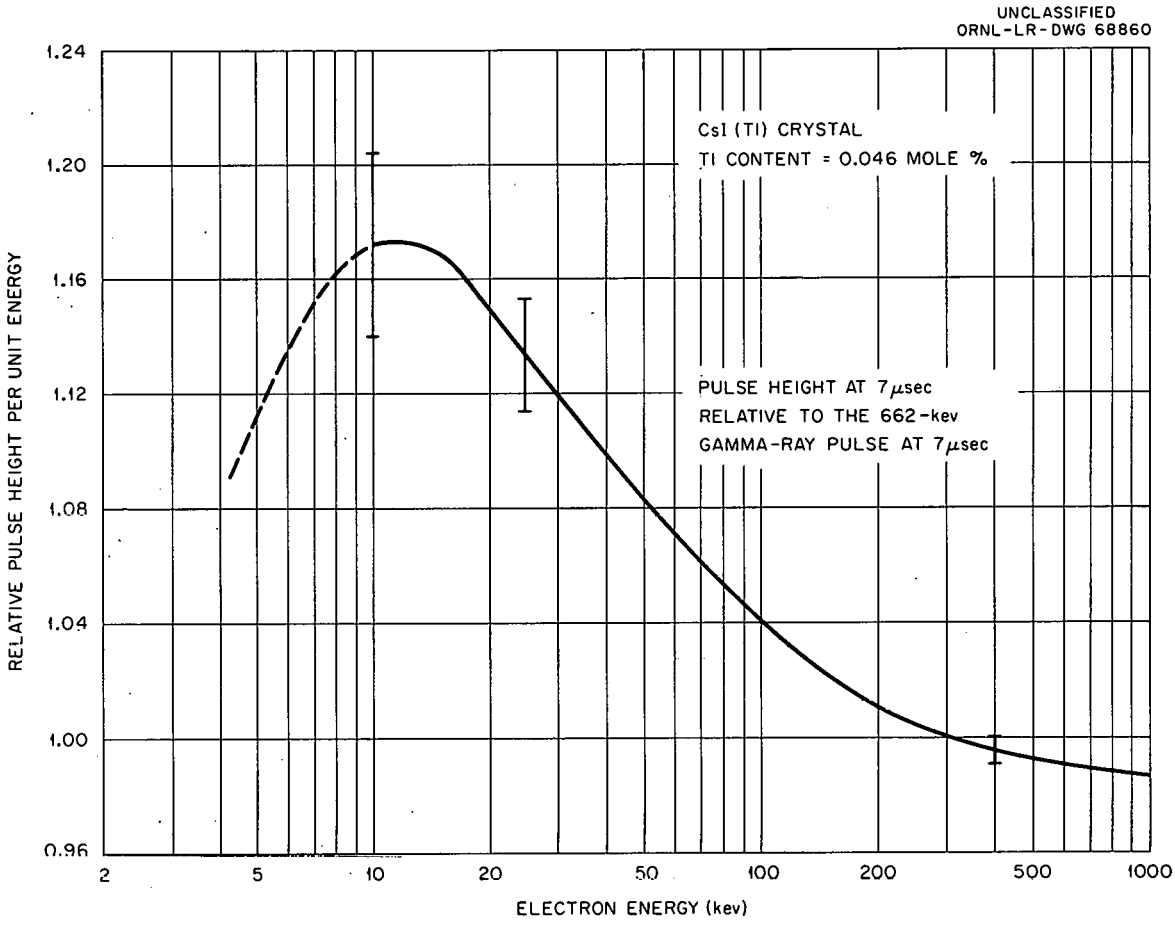

Fig. 23. Derived Value of the Pulse Height Per Unit Energy for Electron Excitation of a CsI(Tℓ) Crystal Having a Thallium Content of 0.046 Mole Per Cent. The dashed curve indicates large uncertainties.

and 23. The dashed line below 10 kev in Figures 22 and 23 indicates that the uncertainties in this region are large.

A calculation was made to estimate the uncertainty introduced in the derived values of the scintillation response of CsI(Tℓ) to electrons because of the simplified model of gamma-ray interactions in the crystal. This calculation was made using the electron response curve derived using the simplified model, and a refined model describing the gamma-ray interactions in the crystal. In this refined model 20 per cent of the photoelectric events occuring for gamma rays having energies above the K-shell absorption energy were assumed to occur with an L-shell electron. For gamma-ray energies below the K-shell absorption energy 20 per cent of the photoelectric events were assumed to occur in a shell having a binding energy of 1 kev. The values of the scintillation response calculated for monoenergetic gamma rays using this latter model were within about 1.5 per cent of the values obtained using the simplified model for the gamma-ray interactions in the crystal. The use of a refined model for the gamma-ray interactions is not justified in the present work because monoenergetic photons were not used in all cases. Furthermore, considerably more data would be needed in the region of the K-shell absorption edge of the elements in the crystal to justify the use of a more refined model.

Figure 24 shows the pulse height per unit energy for gamma rays calculated using the derived electron response curve for the seven-microsecond pulse-analysis time and Equations (11) and (12). The data points in the region of the K-shell absorption energy which are markedly off the calculated curve (approximately 3 per cent) were obtained using

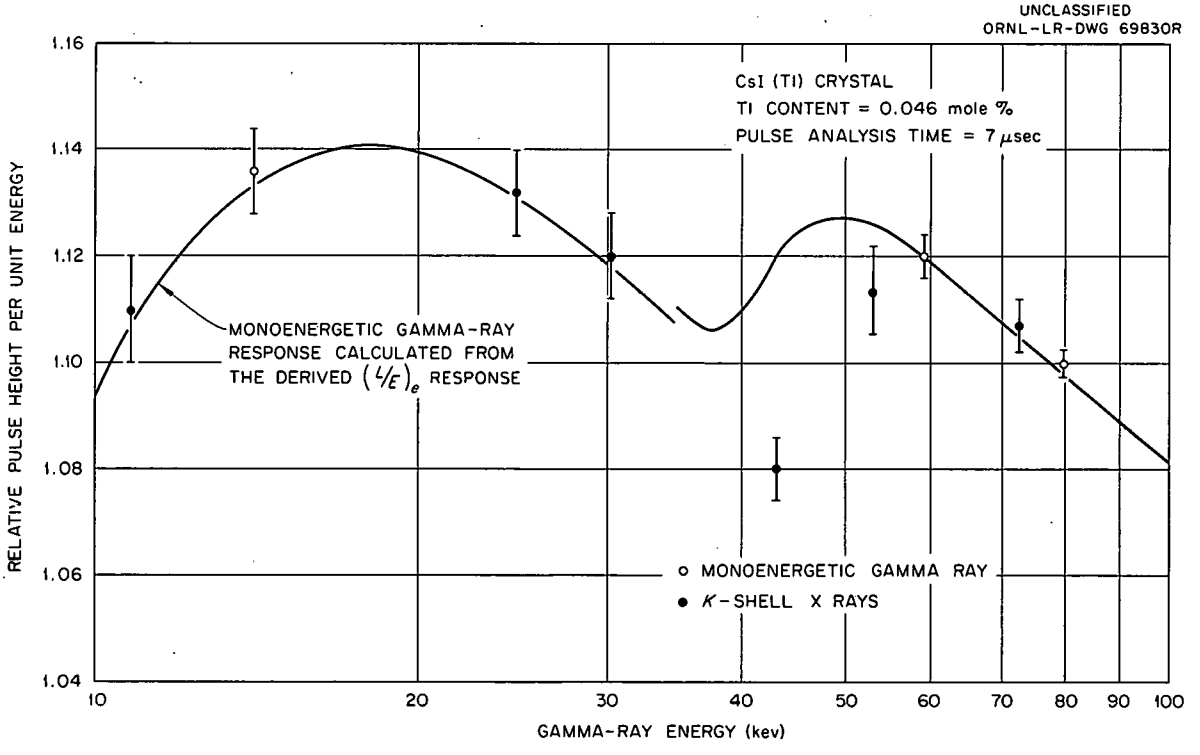

Fig. 24. Pulse Height Per Unit Energy $(L/E)_\gamma$ for Gamma Rays Calculated from the Derived $(L/E)_e$ Curve

K-shell X rays. For gamma rays having energies just above the K-shell absorption energy of the elements of the crystal, the energy of the electron ejected from the K-shell levels is in a region where the uncertainty in the scintillation response is large (see Figure 23). For this reason, it is not surprising to find a lack of complete agreement between the measured values and the derived values of the scintillation response of the crystals to gamma rays in the region of the K-shell absorption energy of the elements in the crystal.

CHAPTER V

COMPARISON OF THE SCINTILLATION MODEL WITH THE EXPERIMENTAL RESULTS

The primary purpose of this experimental program was to test the validity of various aspects of the scintillation model. The scintillation model describes the scintillation process in terms of the dependence of the scintillation efficiency dL/dE upon the stopping power dE/dx (differential energy loss). The scintillation efficiency dL/dE, as calculated from the model, is based upon the total number of photons emitted in a scintillation event.

The time dependence of the scintillation pulse from CsI(Tℓ) has been shown to depend upon the ionization density produced in the crystal by the charged particle.[9] The relative values of the measured pulse heights, for particles producing different ionization densities in the crystal, then depend upon the pulse-analysis time. In the present program measurements were made for pulse-analysis times of one and seven microseconds in order to determine whether there was a gross dependence of the scintillation efficiency upon the pulse-analysis time. The experimental data of Storey, Jack, and Ward[9] show that about 80 per cent of the photons in a scintillation pulse from CsI(Tℓ) produced by gamma rays are emitted in the first seven microseconds. For alpha-particle excitation of CsI(Tℓ) crystals the corresponding quantity is 90 per cent. Thus, in the present work the data obtained from the pulse-height analysis

experiments performed at seven microseconds permit a good comparison to be made with the scintillation model.

The experimental curve of the light output as a function of the energy of the exciting particle must be differentiated to obtain dL/dE in order to make a direct comparison with the theoretical model. Several features of the scintillation model can be investigated, however, without obtaining dL/dE and without considering quantitative values for dE/dx.

One of the major features of the scintillation model is the assumption that dL/dE is a continuously varying function of dE/dx. Since dE/dx is dependent upon the energy of a charged particle, the scintillation model yields the result that the light output is a nonlinear function of the energy of a charged particle. This general result has been observed in the present experimental investigation. The nonlinearity of the light output as a function of the energy of the exciting particle was observed for both pulse-analysis times and for all exciting radiations used in the present work. Figures 9 and 10 show this nonlinear scintillation response of CsI(Tℓ) for excitation by gamma rays, where the pulse height per unit energy is shown as a function of the gamma-ray energy. The data are normalized to unity at 662 kev and it is seen that the shape of the curves is essentially independent of the thallium concentration of the crystal. A proportional response of light output as a function of energy, $L = aE$, where a is a constant, would be a horizontal line in a representation of light output per unit energy as a function of energy. The nonlinearity of light output as a function of energy is most easily detected as a change in the sign of the curvature in a pulse height per unit energy

versus energy representation. The nonproportionality of the light output as a function of energy for CsI(Tℓ) under proton excitation is shown in Figures 13, 14, and 15 for crystals having thallium concentrations of 0.002, 0.046, and 0.17 mole per cent, respectively. The results, for both pulse-analysis times, are shown in the form of pulse height per unit energy versus energy. The nonlinear response of CsI(Tℓ) to protons is most noticable for proton energies less than 3 Mev. The nonlinear response of light output versus energy for alpha particles is clearly shown, for both pulse-analysis times, in Figure 16 for the CsI(Tℓ) crystal having a thallium concentration of 0.046 mole per cent. The data are shown in the form pulse height per unit energy versus energy. The existence of the minimum in the curve of Figure 16 demonstrates the monotonic dependence of the scintillation efficiency on the stopping power for alpha particle excitation of the crystal. On the basis of the scintillation model a minimum in the pulse height per unit energy curve will occur when the energy of the exciting particle passes through the energy region corresponding to the maximum in the stopping power. The minimum of the pulse height per unit energy curve for alpha particles, as shown in Figure 16, does occur in the energy region corresponding to the maximum in the stopping power. Figure 25 shows values of dE/dx for alpha particles as a function of energy of the alpha particle.

Although the light output as a function of energy may be nearly linear in some energy regions for various radiations, it is established that the light output of CsI(Tℓ) as a function of the energy of the charged particle is, in general, nonlinear.

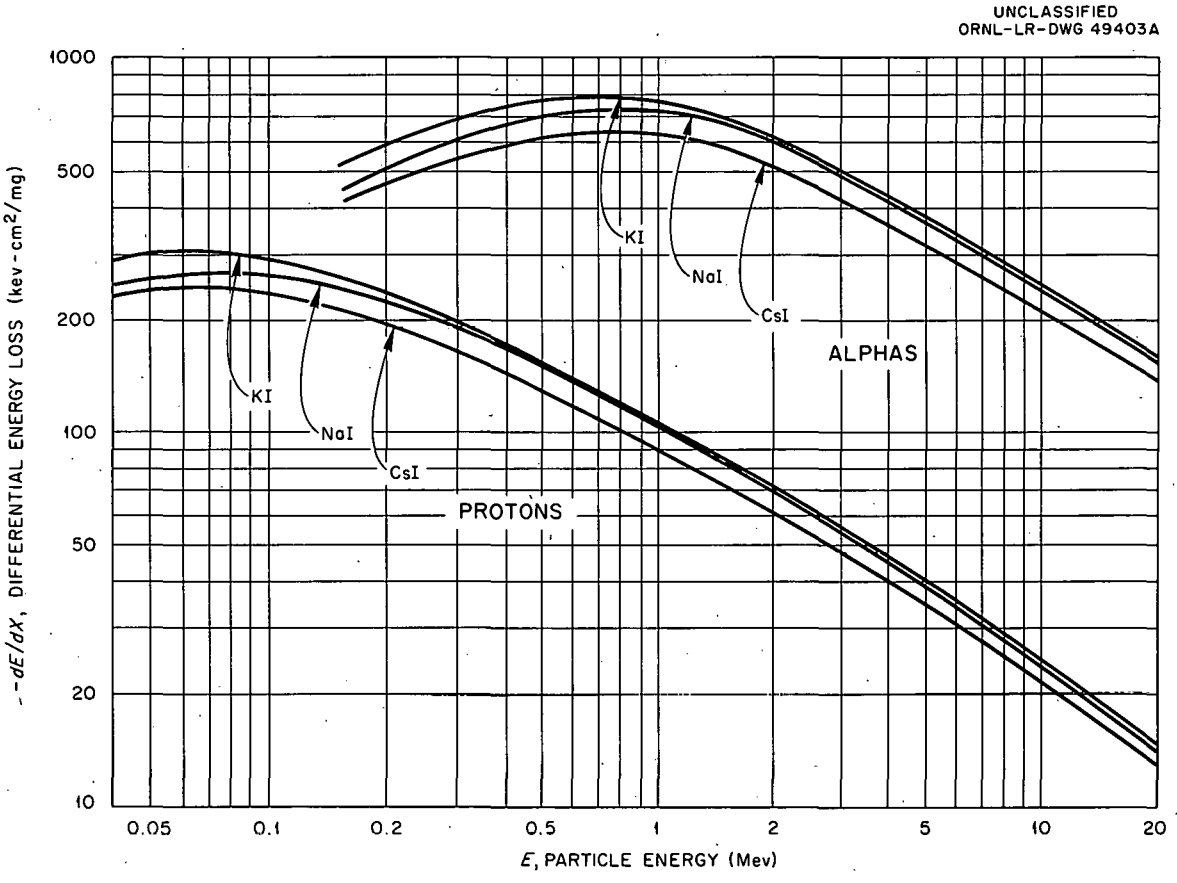

Fig. 25. Values of dE/dx for Protons and Alpha Particles in CsI, KI, and NaI. These values obtained from Reference 1.

The pulse-height response of CsI(Tℓ) to gamma-ray excitation showed nearly the same dependence on the gamma-ray energy for all crystals and for both pulse-analysis times (see Figures 9 and 10). This feature of the gamma-ray scintillation response is described by the scintillation model and is attributed to the recombination of electrons and holes to form excitons, which are assumed to be the energy carriers in the scintillation model. This recombination process was assumed to be an intrinsic property of the crystal and to be independent of the thallium concentration.

The dependence of the scintillation efficiency on the thallium concentration of the crystals as predicted by the scintillation model can be investigated by examining the measured scintillation response of the CsI(Tℓ) crystals for proton excitation. If the thallium-luminescence centers saturate as the dE/dx of the exciting radiation increases, then the pulse height per unit energy versus energy curves for protons should have a different shape for each crystal. An examination of the measured scintillation response of the CsI(Tℓ) crystals to protons, shown in Figures 13, 14, and 15, for crystals having thallium contents of 0.002, 0.046, and 0.17 mole per cent, respectively, shows that there is very little difference in the shape of the light output per unit energy versus energy curves. This observation applies to the data obtained for both pulse-analysis times.

The scintillation response of a CsI(Tℓ) crystal, having a thallium content of 0.31 mole per cent was also measured for excitation by protons. The shape of the pulse height per unit energy versus energy curve for

this crystal was the same as that of the crystal having a thallium content of 0.17 mole per cent.

In order to compare the results of the scintillation experiments directly with the scintillation model the slope dL/dE of the light output versus energy curve must be obtained. The slope of the light output versus energy curve dL/dE was determined from the relation

$$\frac{dL}{dE} \equiv \frac{L}{E} + E \frac{d}{dE} \left(\frac{L}{E}\right). \qquad (14)$$

This technique was used because the light output of CsI(Tℓ) as a function of the energy of the exciting radiation does not deviate strongly from a proportional relation and thus L/E contributes the major portion of dL/dE in Equation (14). The maximum contribution to dL/dE by the term E(d/dE)(L/E) was about 20 per cent and this occured in the analysis of the data for high-energy alpha particles. For the case of protons, the contribution of the term $E \frac{d}{dE}\left(\frac{L}{E}\right)$ to $\frac{dL}{dE}$ ranged from 4 to 12 per cent. The slope of the pulse height per unit energy curve was obtained from the curve L/E by placing a straight edge tangent to the curve at the desired point. The uncertainty in dL/dE introduced by this method of obtaining the slope of the L/E curve was estimated to be about 1 per cent. The uncertainty in drawing a curve of the pulse height per unit energy versus the energy can introduce uncertainties which are larger than 1 per cent. These uncertainties have been considered in the analysis of the experimental data. The values of dL/dE obtained for the various particles were

then plotted as a function of the differential energy loss dE/dx. The values used for dE/dx for alpha particles and protons were those calculated by Murray and Meyer[1] and are shown in Figure 25. The values of dE/dx for electrons were calculated by Meyer[33] using range-energy curves for electrons given in the work of Kanter and Sternglass.[34] The range-energy relations used were obtained by interpolating between curves for the maximum range of electrons in gold and aluminum. There was little dependence of the range, in units of grams per square centimeter, on the atomic number of the element. The values of dE/dx for electrons are shown as a function of the electron energy in Figure 26.

The values of dL/dE for the CsI(Tℓ) crystals derived from the scintillation response experiments are shown as a function of dE/dx in Figures 27 and 28 for pulse-analysis times of one and seven microseconds, respectively. The results for the crystals of different thallium concentrations are shown in each figure. Dashed portions of the curve indicate regions of large uncertainty. The scintillation efficiency dL/dE is seen to vary markedly with the stopping power dE/dx, and the relative values of the scintillation efficiency dL/dE are dependent upon the thallium concentration of the crystals. The curve of dL/dE versus dE/dx for the CsI(Tℓ) crystal having a thallium concentration of 0.046 mole per cent shown in

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[33]A. Meyer, private communication.

[34]H. Kanter and E. J. Sternglass, Phys. Rev. 126, 620 (1962).

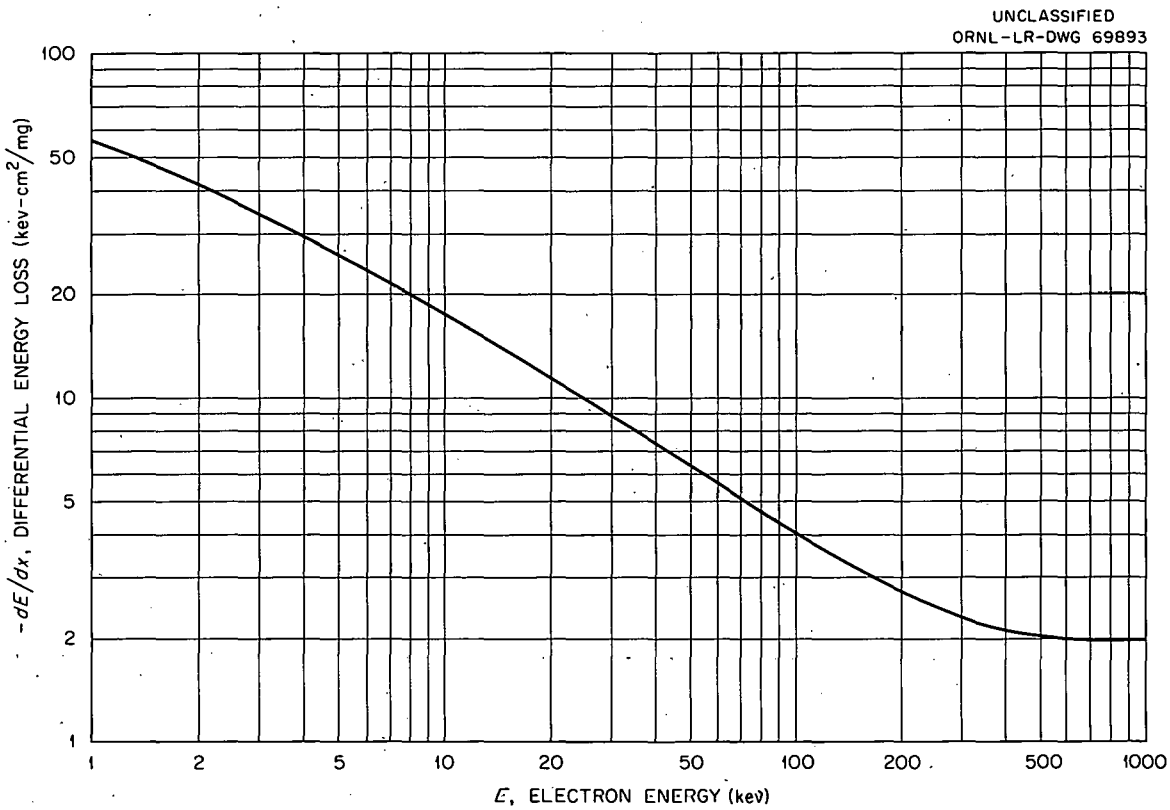

Fig. 26. Values of dE/dx for Electrons in CsI. See Reference 34.

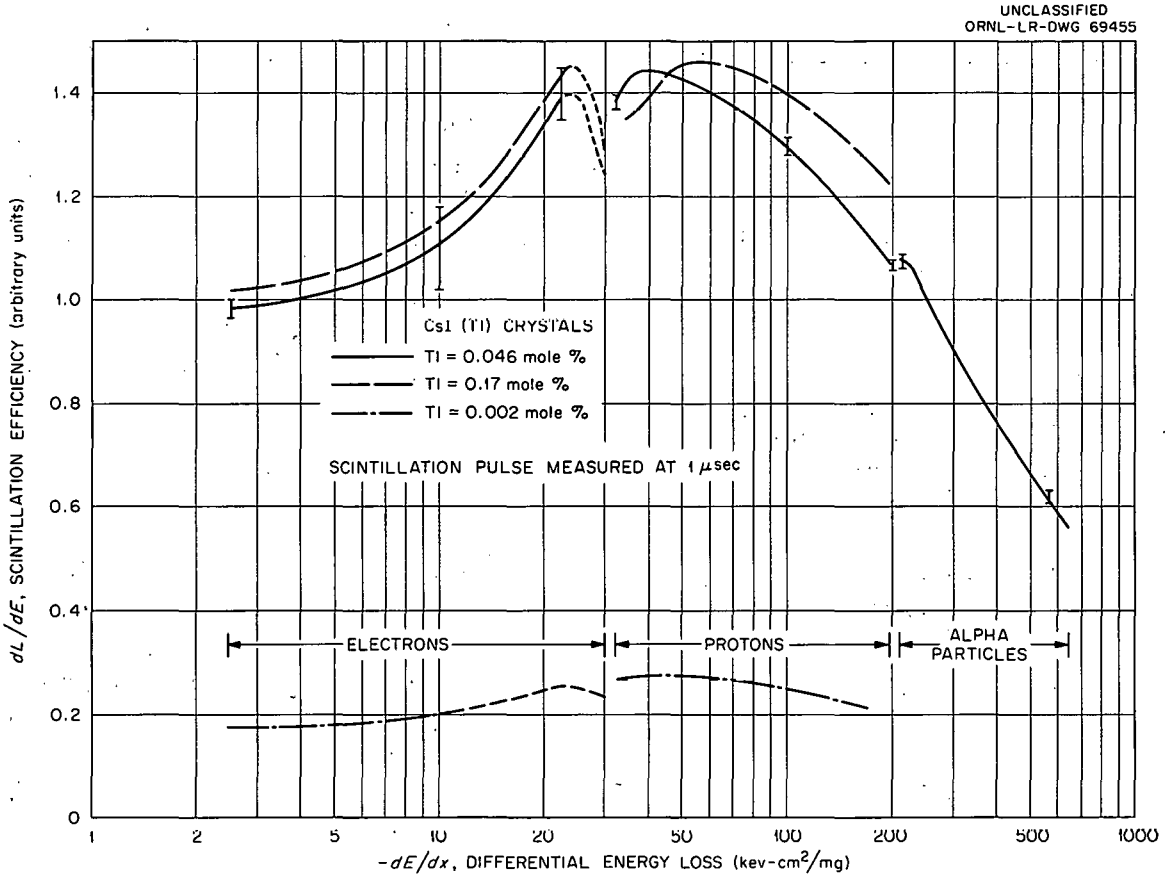

Fig. 27. The Scintillation Efficiency dL/dE as a Function of dE/dx for CsI(Tℓ) Crystals Having Different Thallium Contents. Voltage pulse measured at 1 μsec.

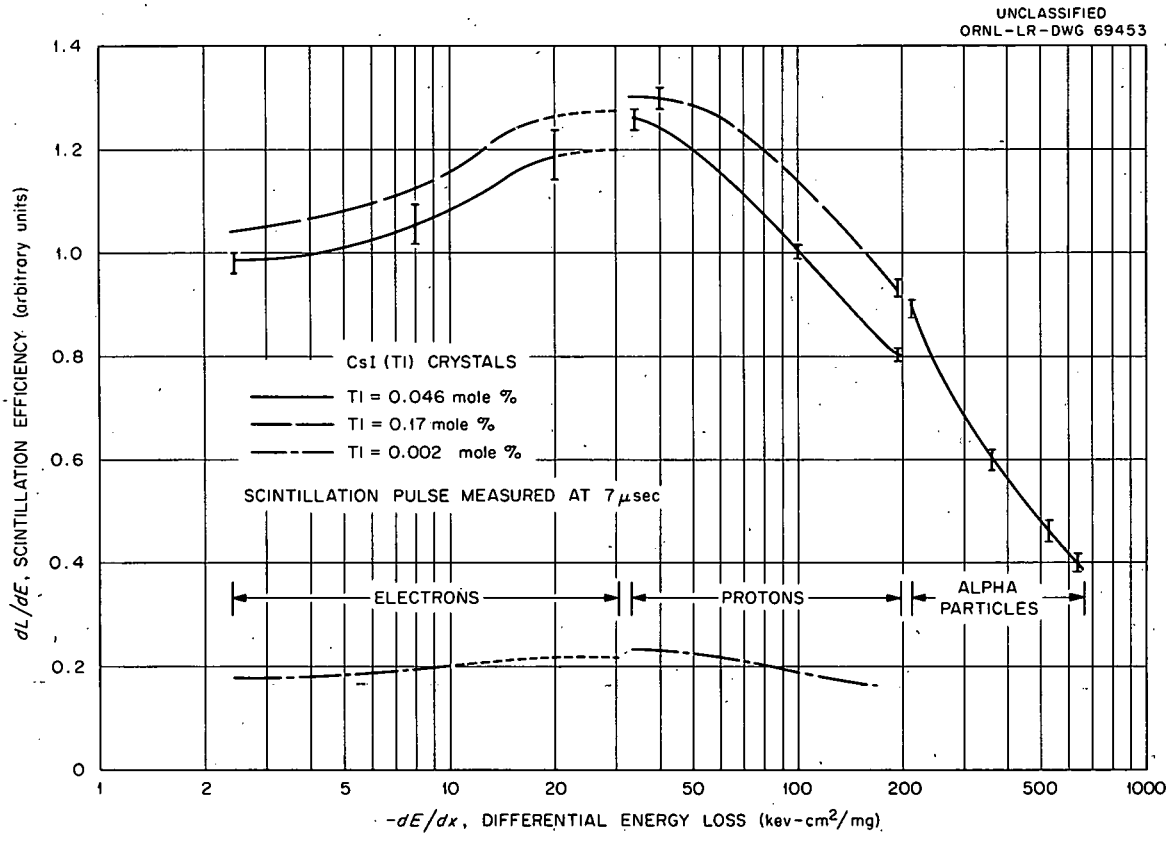

Fig. 28. The Scintillation Efficiency dL/dE as a Function of dE/dx for CsI(Tℓ) Crystals Having Different Thallium Contents. Voltage pulse measured at 7 μsec.

in Figure 28 (seven-microsecond pulse-analysis time) has a discontinuity in the dE/dx region where the portions of the curves for alpha particles and protons join. Although there is uncertainty in the values of dE/dx used it can be seen in Figure 28 that the alpha particle and proton curves have shapes that seem to exclude the discontinuity on this basis. The data observed for a pulse-analysis time of one microsecond in Figure 27 show that the dL/dE versus dE/dx is nearly continuous in the region where the alpha particle and proton portions of the curve meet. The electron and the proton portions of the dL/dE versus dE/dx curves shown in Figures 27 and 28 do join continuously within the uncertainty of the values of dL/dE and dE/dx. The uncertainties in dL/dE are indicated on the figure; uncertainties in dE/dx are estimated to be about 15 per cent for protons and about 50 per cent for electrons in this region of the curve.[33]

The behavior of the dL/dE versus dE/dx curves in the joining region for protons and alpha particles can be explained semi-quantitatively using a method developed by Murray and Meyer which considers the effect of energetic secondary electrons (delta rays) produced by charged particles traversing the crystal.[35] Some of these delta rays have sufficient energy to escape the region of high ionization produced by the primary particle and these delta rays produce light with an efficiency characteristic of low-energy electrons. For alpha particles and protons having the same dE/dx, the alpha particle will produce higher energy delta rays than

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[35]R. B. Murray and A. Meyer, IRE Trans. on Nuclear Sci. NS-9, 33 (1962).

protons will; thus there will be more contribution to the luminescence from delta rays for alpha particles than for protons. A calculation based upon the delta-ray model of Murray and Meyer showed that about 7 per cent of dL/dE for 10-Mev alpha particles was due to delta rays. The magnitude of the discontinuity between the value of dL/dE for protons and alpha particles is about 12 per cent (see curve in Figure 28 corresponding to a pulse-analysis time of seven microseconds) at a dE/dx of 210 kev-cm^2/mg. The uncertainty in dL/dE is about 4 per cent for both the proton curve and the alpha-particle curve. The discontinuity between the alpha particle and proton values of dL/dE at about 210 kev-cm^2/mg for a pulse-analysis time of one microsecond (see Figure 27) is not as pronounced as for the pulse-analysis time of seven microseconds. This result is expected since a greater fraction of the emitted light appears in the long decay component of CsI(Tℓ) for excitation by electrons than for alpha particles. On this basis the difference in dL/dE for protons and alpha particles for a pulse-analysis time of one microsecond should be about one-half that calculated using the delta-ray model which is based on total light emission. This difference, which is about 3.5 per cent of dL/dE, is less than the expected uncertainty in comparing values of dL/dE for protons and alpha particles. A discontinuity in dL/dE is not expected in the region of dE/dx in which the proton and electron portions of the curve overlap because the scintillation efficiency of the primary particle and delta rays are essentially the same in this region. The results of this experimental program are, therefore, considered to be in

good agreement with the scintillation model, as modified by the effect of delta rays, on the continuity of dL/dE versus dE/dx.

The dependence of the shape of the curve dL/dE versus dE/dx on the thallium content of the crystals is shown in Figure 29 for a pulse-analysis time of seven microseconds. Figure 29 (pulse-analysis time of seven microseconds) shows values of dL/dE obtained for CsI(Tℓ) crystals having thallium contents of 0.002, 0.046, 0.17, and 0.31 mole per cent. The curves for the different thallium concentrations are normalized to the electron portion of the curve. The shape of the dL/dE versus dE/dx is dependent upon the pulse-analysis time as expected, since the time dependence of the scintillation pulse is a function of dE/dx. The curves of dL/dE versus dE/dx are not the same for each crystal, but the quantitative dependence of the curves on the thallium content of the crystals as predicted by the scintillation model is not observed.

The total light emitted in the scintillation pulse was calculated using Equation (2), which gives the ratio of the light emitted in the first seven microseconds to the total light emitted in the scintillation pulse. The values of the parameters appearing in Equation (2) were taken from the work of Storey, Jack, and Ward, and the parameters were assumed to be continuous functions of the stopping power.[9] The total light emitted in the scintillation pulse in the two components described in Equation (2) is about 10 to 20 per cent greater than the light emitted in the first seven microseconds of the scintillation pulse, and thus the uncertainty introduced by using Equation (2) to obtain the total light in these two components should not be large. Figure 30 shows dL/dE versus dE/dx for

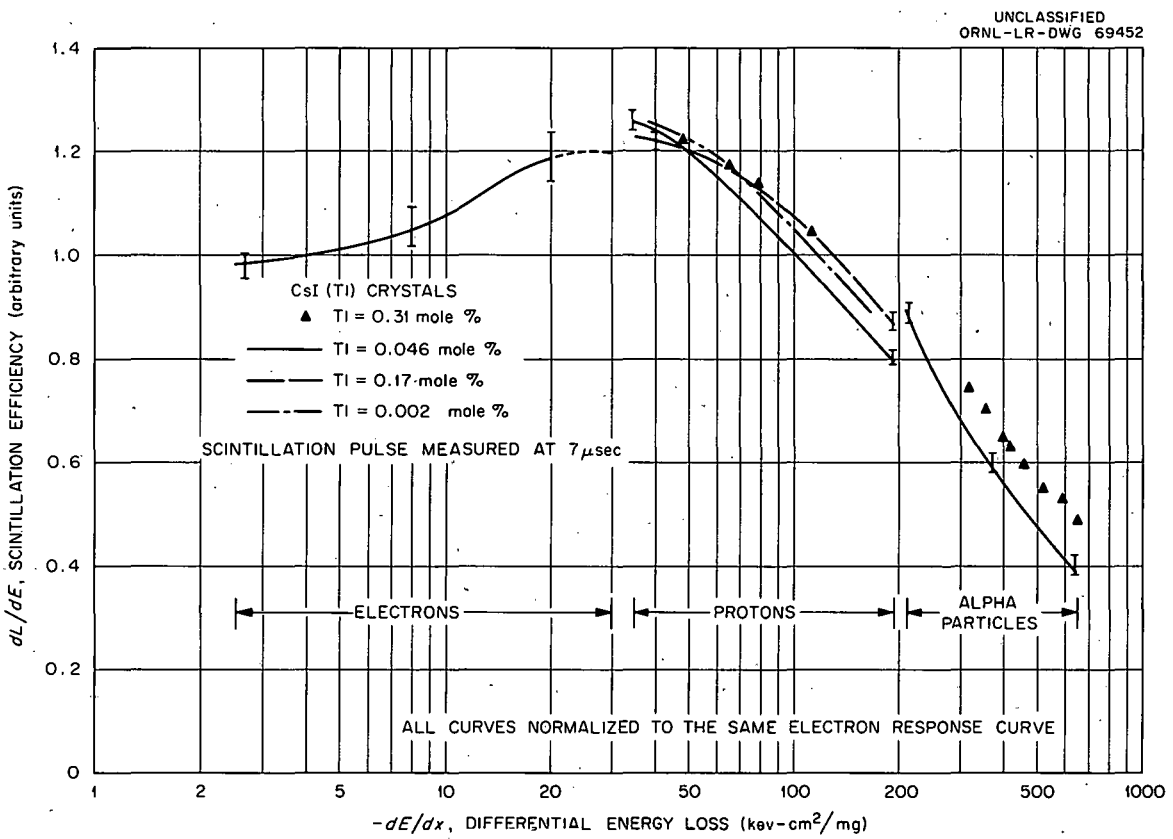

Fig. 29. The Scintillation Efficiency dL/dE Versus dE/dx for a CsI(Tℓ) Crystal Having Different Thallium Contents. The curves are normalized to the same electron response curve.

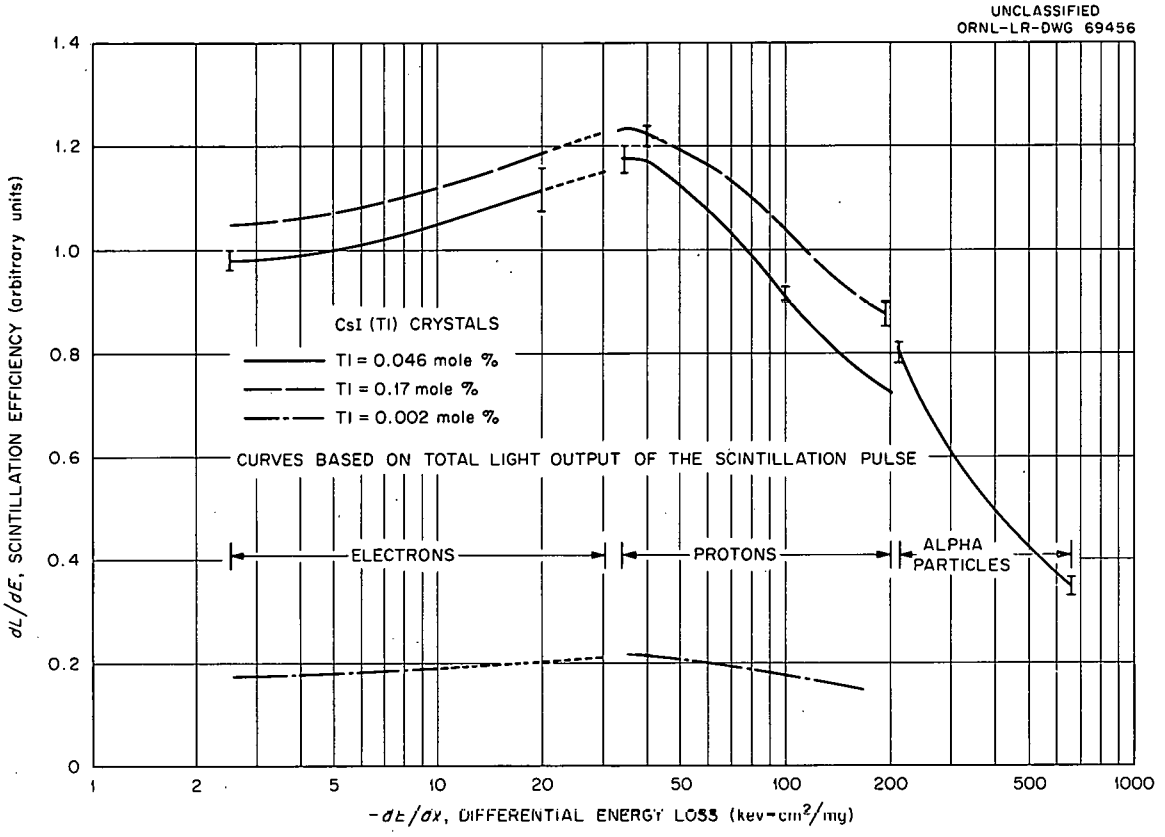

Fig. 30. Estimated Values of the Scintillation Efficiency dL/dE Versus dE/dx for CsI(Tℓ) Crystals Having Various Thallium Contents. The values of dL/dE are based on an estimate of the relative voltage pulses at t = ∞.

the various crystals based on the total light output of the scintillation pulse. Figure 31 shows a comparison of the curves dL/dE versus dE/dx as derived experimentally and as calculated from the scintillation model for CsI(Tℓ) crystals.* The curves are normalized to the electron portion of the curves at low values of dE/dx. The experimental curves clearly do not have the dependence upon thallium concentrations as predicted by the scintillation model, and it may be concluded that the decline of the scintillation efficiency over the range of dE/dx covered is not caused by the saturation of thallium-luminescence centers as described by the scintillation model.

This experimental program has demonstrated that within the experimental uncertainties the scintillation efficiency is a continuous function of the stopping power, over the range of dE/dx investigated, when the effects of delta rays are considered. [This leads to the conclusion that the light output of CsI(Tℓ) is in general a nonlinear function of the energy of the exciting particle over the range of the present experiments since dE/dx is a continuously varying function of energy.] Experiments performed on CsI(Tℓ) using heavy ions show that the nonlinear response of CsI(Tℓ) continues for values of dE/dx greater than that covered in the present experimental program.[36,37]

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*The theoretical curves were calculated by A. Meyer of the Oak Ridge National Laboratory.

[36]E. Newman and F. E. Steigert, Phys. Rev. 118, 1575 (1960).

[37]E. Newman, A. M. Smith, and F. E. Steigert, Phys. Rev. 122, 1550 (1961).

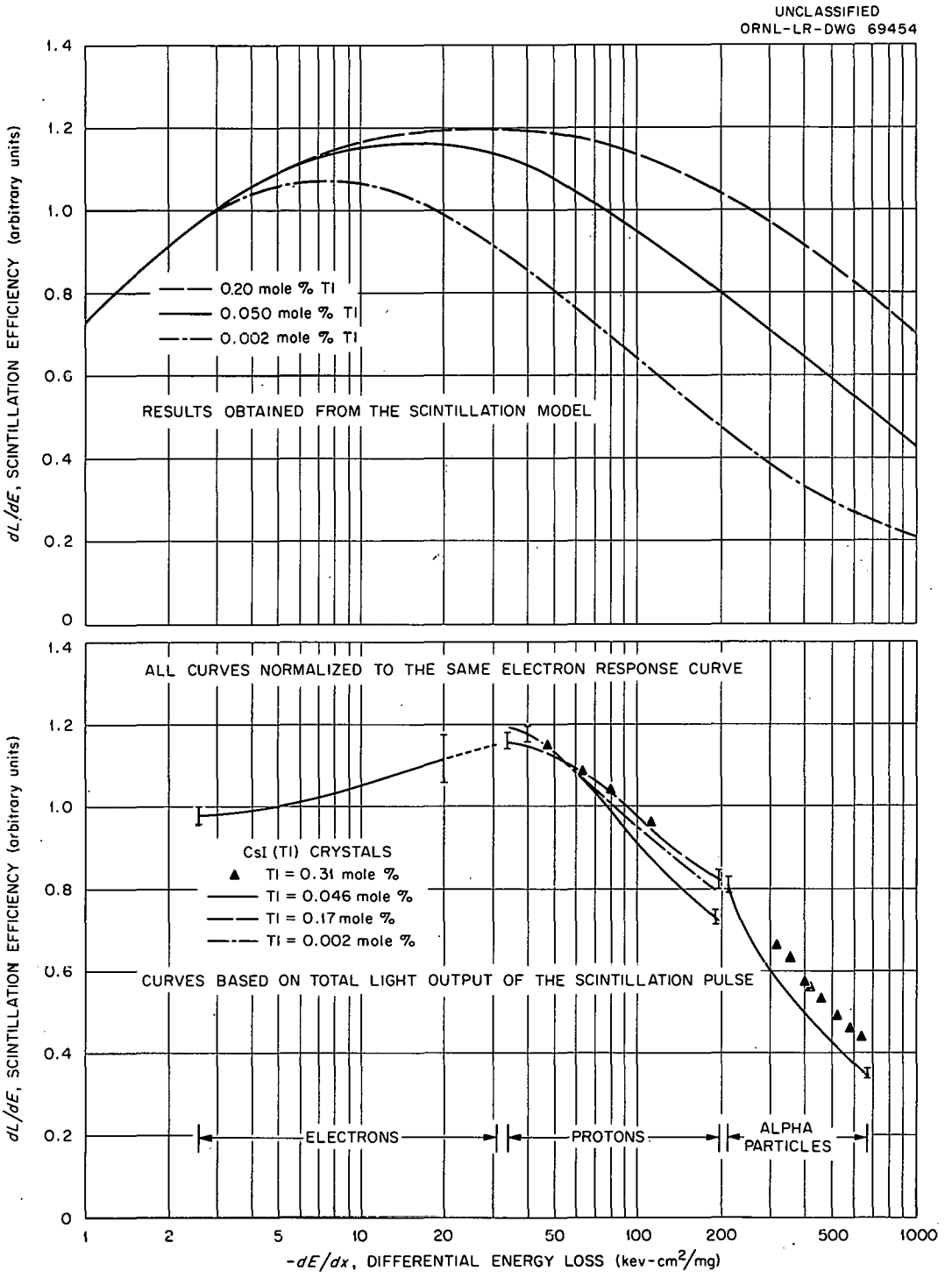

Fig. 31. Comparison of the curves of dL/dE Versus dE/dx Obtained from the Experiments with the Values Predicted by the Scintillation Model.

CHAPTER VI

DISCUSSION OF THE EMISSION SPECTRUM OF CsI(Tℓ)

The emission spectra of CsI(Tℓ) crystals having different thallium concentrations were measured over the wavelength range extending from 3000A to 7500A for excitation by X rays, protons, and alpha particles. A broad emission spectrum was observed in all cases with the maximum intensity occurring at about 5700A. Figure 20 shows the emission spectra for a CsI(Tℓ) crystal having a thallium content of 0.17 mole per cent for excitation by 1.4- and 4.4-Mev protons and 2-Mev alpha particles. All of the curves showing the emission spectrum of CsI(Tℓ) are normalized to the same area. All of the emission spectra shown in Figure 20 exhibit the same features; the maximum in the intensity occurs at about 5700A, and the intensity at 7000A is about 20 per cent of the maximum value. The emission spectra curves have a shoulder in the vicinity of 4000A, and below 4000A the intensity drops rapidly. Figure 19 shows the emission spectra of CsI(Tℓ) having a thallium content of 0.046 mole per cent under excitation by 50-kev X rays, 1.4-Mev protons, and 2-Mev alpha particles. For this crystal an emission band was observed at about 3300A for excitation by alpha particles. Figure 18 shows the emission spectra of a CsI(Tℓ) crystal having a thallium content of 0.002 mole per cent for excitation by 250-kev X rays, 1.4-Mev protons, and 8.7-Mev alpha particles. For this crystal the band at about 3300A is apparent for all exciting radiations except X rays; there appears to be an emission band at 3300A for the X rays, but its intensity is relatively weak. The relative intensity of the

3300A band increases as the ionization density produced in the crystal increases. Figure 21 shows the emission spectra for a pure CsI crystal (no thallium intentionally added) and for CsI(Tℓ) crystals having thallium contents of 0.002, 0.046, and 0.17 mole per cent for excitation by 4.4-Mev protons. The relative intensity of the 3300A band increases as the thallium content decreases but the emission spectrum above 4000A is nearly the same for all the thallium-activated crystals. All of the emission spectra for the thallium-activated crystals exhibit a shoulder in the vicinity of 4000A. The "pure" (no thallium) crystal exhibits three distinct bands, one at 3300A, another at about 4700A, and one at about 6500A. (The emission band at 6500A may be due to second order diffraction of the light from the 3300A band.)

The emission spectra of pure and thallium-activated CsI crystals were measured by Morgenshtern.[12,14] The pure crystals used were prepared from aqueous solutions and also grown from the melt. Three emission bands were observed for excitation by gamma rays, one in the vicinity of 3300A, another at 4000A, and the thallium band at about 5800A. The pure crystals grown from the aqueous solutions exhibited only the 3300A band, while the crystals grown from the melt always had an emission band at 4000A and in certain cases there was an emission band at 3300A. Morgenshtern[14] suggested that the 4000A emission band might be due to vacancies in the crystal lattice.

The emission spectrum of CsI(Tℓ) in the wavelength region from 4000A to 6000A was measured by Lagu and Thosar for excitation by ultraviolet light and by gamma rays.[29] A broad spectrum was found with peaks

occuring at 4400A, 4750A, 5650A, and 5950A for both gamma-ray excitation and for excitation by a broad band of ultraviolet light extending from 2300A to 3000A. For excitation by ultraviolet light above 3000A peaks at 5660A and 6050A were observed.

Hrehuss also observed that the emission spectrum of CsI(Tℓ) depended upon the charged particle exciting the crystal.[13] He measured the emission spectrum in the range from about 4200A to 5700A and observed that the relative intensity of light in the region below 5000A increased as the ionization density produced by the charged particles increased. The relative amount of light emitted below 5000A was a factor of about two greater for excitation by electrons than for alpha particles. This type of behavior was noted in the present work for wavelengths less than about 4000A. The current density of charged particles exciting the crystals was about 10^3 greater in the experiments of Hrehuss[13] than that used in the present work, and it is possible that effects due to the different excitation rates were responsible for the differences in the measurements.

In the current work a difference in the emission spectrum of a given CsI(Tℓ) crystal was noted as the time of irradiation of the crystal increased. As the irradiation time increased for alpha-particle excitation, the relative intensity of the 3300A band increased for the CsI(Tℓ) crystals having thallium concentrations of 0.046 and 0.002 mole per cent. There was a small increase in the relative intensity of the spectrum in the 4000A region for the CsI(Tℓ) crystals having a thallium concentration of 0.002 mole per cent. It could not be determined from the available

data whether the relative increase in the 3300A band was due to an increase in absolute yield of that band or a decrease in the yield of the band associated with the thallium.

The broad emission band observed at 4700A for the "pure" CsI crystal is possibly caused by impurities in the crystal since Morgenshtern reported that in CsI crystals grown from aqueous solutions using purified cesium iodide only the 3300A band was observed. The shoulder at 4000A in the emission spectrum of CsI(Tℓ) observed in the present work may be due to lattice vacancies as suggested by Morgenshtern.[14]

The absorption spectrum of CsI(Tℓ) shows a peak due to the presence of thallium at about 2990A; the tail of the absorption peak extends to about 3400A.[30] The luminescence band associated with the thallium activator is stimulated by excitation with ultraviolet light for a wavelength range extending from less than 2000A to about 3500A. The fact that the relative intensity of the 3300A emission band is a function of the thallium concentration is certainly due in part to the absorption of the 3300A band light by the thallium centers in the crystal, and the resultant emission of light in the thallium-activator band. It is also possible that the thallium centers in the crystal compete with the luminescence centers giving rise to the 3300A emission band for excitation and that very little light is emitted in the 3300A band in crystals having large thallium concentrations. The fact that the relative intensity of the 3300A emission band increases as the ionization density produced by the charged particle increases indicates that the thallium centers and the 3300A emission band centers compete for excitation. The observed

dependence of the relative intensity of the 3300A emission on the ionization density and the thallium concentration of the crystal can be explained by a saturation process of the type described by the scintillation model. Another possible explanation of the dependence of the relative intensity of the 3300A emission band is that the centers which are responsible for the 3300A emission band are created by the charged particle in the ionization process and that the density of these centers increases as the ionization density increases. This mechanism is discussed in more detail in Chapter VII.

The absorption coefficients of the crystals, for ultraviolet light, used in the present experimental program are not known. A relative measurement of the ultraviolet light transmission of the "pure" CsI crystal and the CsI(Tℓ) crystals having thallium contents of 0.002 and 0.17 mole per cent was made.* Absorption peaks were noted in the CsI(Tℓ) crystal having a thallium content of 0.002 mole per cent, which corresponded to the absorption peaks in CsI(Tℓ) observed by Forro.[30] These experiments also indicated that the transmission of ultraviolet light in the wavelength region of 3300A by the thallium-activated crystals was small. For example, at a wavelength of 3250A less than 1 per cent of the incident light was transmitted by the 2-mm thick CsI(Tℓ) crystal, which had a thallium content of 0.17 mole per cent. The corresponding fraction of the incident light (3250A) transmitted by the crystal having a thallium

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*These measurements were made by D. A. Costanzo of the Oak Ridge National Laboratory.

content of 0.002 mole per cent was about 50 per cent. These values giving the per cent transmission are approximate and are not intended to represent the exact fraction of the 3300A emission band which is absorbed by these crystals.

CHAPTER VII

DISCUSSION AND CONCLUSIONS

The results of this experimental investigation have shown that the scintillation efficiency of the thallium-activator luminescence in CsI(Tℓ) is a continually varying function of dE/dx as assumed in the scintillation model. The scintillation model predicted that the decline in dL/dE at large values of dE/dx was due to a saturation of thallium sites as the density of energy carriers increased. The results have shown that the saturation phenomenon as described by the scintillation model does not occur in CsI(Tℓ). The present results have not demonstrated, however, that a saturation of thallium-luminescence centers does not occur to some extent. The fact that there is an emission band characteristic of pure crystals of CsI which can stimulate the emission of light in the thallium-activator band complicates the interpretation of the experimental data on the scintillation response in terms of a saturation process.

Since the decline in dL/dE at large values of dE/dx is not adequately described by the saturation of thallium centers, as described by the scintillation model, a review of some possible causes of this phenomenon is in order. Some effects which might result in a decrease in dL/dE as dE/dx increases are: (1) a change in the emission spectrum of the scintillator, (2) effects due to the time of the pulse-height analysis, (3) instrumental effects, (4) a "dead layer" for scintillations at the surface of the crystal, and (5) ionization quenching effects.

In connection with item (1) the present experimental program has shown that the decrease in dL/dE for thallium-activator luminescence at large values of dE/dx is not due to a change in the emission spectrum in CsI(Tℓ).

With respect to item (2), the results of the current program have also shown that the time dependence of the scintillation pulse is not responsible for the decline of dL/dE at large values of dE/dx. This statement does not apply to the long term phosphorescence components having decay times which are long compared to seven microseconds.

With respect to item (3), Engelkemeir[21] demonstrated that the electronic system was not responsible for the nonlinear response of NaI(Tℓ) to gamma rays by using a pulse generator (in the same manner as used in the present work) to calibrate the electronic system. He also demonstrated that the gain of the photomultiplier tube was not the cause of the nonlinear response of NaI(Tℓ) to gamma rays by changing the gain of the tube by a factor of twenty-five. Managan[22] observed the nonlinear response of NaI(Tℓ) for gamma-ray excitation, and used a pulse attenuator to demonstrate that the electronic system was not responsible for the observed nonlinear response. In addition, he proved that the photomultiplier was not responsible for the observed nonlinearity by using a light pulser to check the response of the photomultiplier tube. In the present work the pulse heights, corresponding to excitation of the crystal by a given particle, were measured relative to the pulse height of the 662-kev gamma-ray standard. In addition, the electronic system was calibrated after each run using a precision pulser in connection with a laboratory potentiometer.

With respect to item (4), a variety of reasons can be given to demonstrate that a "dead layer" for scintillation does not produce the low scintillation efficiency observed at large dE/dx. The experiments of Taylor and his associates on the relative scintillation response of NaI(Tℓ) to electrons in the energy region from 0.5 to 624 kev and to alpha particles for an energy range from 4 to 21 Mev show that a "dead layer" for scintillations is not responsible for the high value of dL/dE for electrons compared to that for alpha particles.[38] The scintillation efficiency for excitation by electrons was approximately constant over the energy range from about 1 kev to 624 kev and the magnitude of the scintillation efficiency was about twice that for alpha particles having energies of 8 Mev. The range of an 8-Mev alpha particle in sodium iodide crystals is about 10^3 times that of 1-kev electrons; this fact eliminates the possibility that a "dead layer" for scintillations produces the lower scintillation efficiency for alpha particles. This conclusion is substantiated by the experiments of der Mateosian and Yuan in which an alpha-particle-emitting substance was incorporated in a NaI(Tℓ) crystal in order to compare the scintillation response of the crystal for excitation by an alpha particle both internal to external to the crystal.[39] The observed difference of about 5 per cent on the relative response to alpha particles of the same energy was not great enough to account for the decline in dL/dE at large values of dE/dx. Experiments were performed by

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[38]C. J. Taylor, W. K. Jentschke, M. E. Remley, F. S. Eby, and P. G. Kruger, Phys. Rev. 84, 1034 (1951).

[39]E. der Mateosian and Luke C. L. Yuan, Phys. Rev. 90, 868 (1953).

der Mateosian, McKeown, and Muehlhause to compare the relative values of the pulse height per unit energy in NaI(Tℓ) for excitation by electrons and alpha particles.[40] The alpha-particle sources were incorporated in the NaI(Tℓ) crystal. At room temperature the pulse height per unit energy for excitation by alpha particles was about 70 per cent of that for excitation by electrons. This demonstrates that a surface "dead layer" is not responsible for the observed decrease in dL/dE at large values of dE/dx. It is possible that a larger concentration of crystalline defects are present near the surface of the crystal than at the interior of the crystal, and that these defects do have a small effect upon the scintillation response of the crystal to various charged particles. The magnitude of this effect must be less than about 5 per cent for the case of excitation with alpha particles.[39]

It is concluded that a "dead layer" for scintillations does not produce the decline in dL/dE observed at large values of dE/dx. This conclusion is further substantiated by experiments on the interaction of fast neutrons in NaI(Tℓ) and Li^6I(Eu) crystals.[41,42] In these experiments the reaction products are produced in the interior of the crystal, but the scintillation efficiency to high dE/dx particles is low.

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[40]E. der Mateosian, M. McKeown, and C. O. Muehlhause, Phys. Rev. 101, 967 (1956).

[41]E. N. Shipley, G. E. Owen and L. Madansky, Rev. Sci. Instr. 30, 604 (1959).

[42]R. B. Murray, Nucl. Instr. 2, 237 (1958).

Item (5) concerns the concept of an ionization quenching effect. The decrease in dL/dE versus dE/dx in organic scintillators was attributed to ionization quenching effects by Birks.[43] In this concept the density of traps for energy carriers increases as the ionization density increases. The application of this model to an activated inorganic crystal results in a relation of the form $dL/dE = A/[1 + (B/N[T\ell])(dE/dx)]$, where A and B are constants. Examination of this function shows that the predicted shape of the dL/dE versus dE/dx curve depends on the thallium concentration of the crystal in a manner similar to that expected from the scintillation model. Since the dependence of the shape of dL/dE versus dE/dx observed in the present work was small, it can be concluded that a simple ionization quenching model does not adequately describe the variation of dL/dE versus dE/dx. Since none of the items considered in this work seem to account for the observed values of dL/dE at large dE/dx, it is necessary to examine further some of the details of the scintillation process in the hope of understanding the observed behavior of dL/dE for large values of the ionization density.

One phenomenon which was observed in the present experimental program may aid in the understanding of certain aspects of the scintillation process. The relative intensity of the 3300A emission band, with respect to the thallium-activator band, was observed to be dependent upon the thallium concentration of the crystal and the ionization density produced in the crystal by the charged particle (see Figures 18-21). As

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[43]J. B. Birks, Phys. Rev. 84, 364 (1951).

the thallium content of the crystals decreased the relative intensity of the 3300A emission band increased, and as the ionization density produced in the crystal increased the relative intensity of the 3300A emission band increased. This result indicates that there is competition between thallium-activator luminescence centers and the luminescence centers responsible for the 3300A emission band in CsI(Tℓ) and that this competition depends upon the ionization density.

The light emitted in the 3300A band in CsI(Tℓ) crystals is attenuated by an absorption peak in CsI(Tℓ) which is due to the presence of thallium in the crystal.[30] In addition, at room temperature thallium-activator luminescence is stimulated by the absorption of light in this absorption peak of CsI(Tℓ).[4,12,29] Thus, the relative emitted intensity of the 3300A emission band with respect to the thallium-activator luminescence band is larger than the relative value of the emergent intensity measured in the present experiments. Measurements of the transmission of light in the region of 3300A by the 0.2-cm-thick CsI(Tℓ) crystal having a thallium content of 0.002 mole per cent showed that about 50 per cent of the incident light was absorbed. This value was based on a comparison with a "pure" CsI crystal. The existence of the absorption of light in the region of 3300A in the crystals due to the presence of thallium was definitely established in these experiments. The surfaces of the crystals were polished, but undoubtedly the surface reflection was different for each crystal. Thus, a generous uncertainty of at least a factor of two must be assigned to the attenuation factors. For the 0.2-cm-thick CsI(Tℓ) crystal having a thallium content of 0.17 mole per cent, the fraction of

the incident light in the region of 3300A which was transmitted was less than 1 per cent. The fact that the 3300A emission band was observed for alpha-particle excitation of the crystal which had a thallium content of 0.046 mole per cent (see Figure 19) indicates that the emitted intensity of this band may have been an order of magnitude greater than the observed emergent intensity. On this basis about 5 per cent of the photon emission was in the 3300A band. Since the emission band at about 3300A in CsI overlaps the absorption band of thallium-luminescence centers in the crystal, it is possible that sensitized luminescence occurs, as well as optical excitation of the thallium-activator band. The probability of the transfer of energy in a radiationless transition from one center to another in sensitized luminescence increases as the overlapping of the emission spectrum of the exciting center and the absorption spectrum of the excited center increases.[44] A knowledge of the origin of the 3300A band in CsI crystals should aid in understanding the mechanisms of the scintillation process.

The 3300A emission band of CsI is most intense for crystals which have been grown from aqueous solutions of highly purified cesium iodide.[14] At the temperature of liquid nitrogen the luminescence yield of the 3300A band in these CsI crystals grown from aqueous solutions is of order of 50 per cent (in terms of energy conversion) for excitation of the crystal by Co^{60} gamma rays. At room temperature the luminescence yield is about a factor of ten less than the yield at the temperature of liquid nitrogen.[14]

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[44]D. L. Dexter, J. Chem. Phys. 21, 836 (1953).

The luminescence yield of the 3300A band in CsI crystals grown by the Stockbarger method was not as high as the yield of the crystals grown from an aqueous solution. In some crystals grown from the melt the 3300A emission band was not observed at all. The 3300A band was observed, in the present experimental program, in CsI(Tℓ) excited by charged particles. It is interesting to note that, for excitation of the crystal with 250-kev X rays, an emission band at 3300A was not observed even for the CsI(Tℓ) crystal which had a thallium content of 0.002 mole per cent (see Figure 18). For excitation of this crystal (thallium content of 0.002 mole per cent) with 50-kev X rays, a very small emission peak was observed in the region of 3300A; this emission spectrum is not shown. For CsI crystals which exhibit an emission band other than the 3300A emission band, the intensity of the 3300A band is negligibly small for gamma-ray excitation of the crystals at room temperature.

Morgenshtern observed an emission band at 3300A, 4000A, and about 5800A in a CsI(Tℓ) crystal, at the temperature of liquid nitrogen, excited by gamma rays; the thallium content of the crystal was about 10^{-4} mole per cent.[14] At room temperature the luminescence yield of the thallium band was about the same as at the temperature of liquid nitrogen, but the 3300A emission band was not observed. These data on the 3300A emission band indicate that there is a nonradiative transition associated with the center responsible for the 3300A band, and as the temperature increases the probability of the nonradiative transition increases. Since the 3300A emission band was observed, in the present work, for excitation of the CsI(Tℓ) crystals at room temperature by charged particles, it is possible

that a nonradiative process is taking place with a transition probability of order ten times that of the radiative transition probability. The center associated with the 3300A emission band seems to compete with the thallium and may be responsible for the decline in dL/dE at large values of dE/dx. As stated previously, the relative intensity of the 3300A emission band was observed to increase with respect to the thallium-luminescence band as the ionization density increased. Since the light output per unit energy of CsI(Tℓ) decreases as the ionization density produced by the charged particle increases, the increase in intensity of the 3300A band as the dE/dx increases suggests that the relatively low values of dL/dE at large values of dE/dx are associated with the center giving rise to the 3300A band. The magnitude of this competition increases as the ionization density in the crystal increases.

A center which apparently has the features attributed to the center associated with 3300A emission band is a self-trapped hole. There are centers in alkali halide crystals which have been identified as halide molecule ions of the type X_2^-. In particular, an I_2^- molecule ion has been identified in a KI crystal at low temperatures.[45] The identification of the I_2^- molecule ion in KI was based upon an investigation of the optical and paramagnetic resonance spectra of alkali halide crystals which contained electron traps. These centers have been observed after X ray irradiation of the alkali halide crystals at the temperature of liquid

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[45]C. J. Delbecq, W. Hayes, and P. H. Yuster, Phys. Rev. 121 1043 (1961).

nitrogen. The rate of formation of these X_2^- centers can be increased by orders of magnitude by adding an electron trap, such as Tl^+, to the crystal.[45] The lifetime of these X_2^- molecule ions depends upon the temperature; at a sufficiently low temperature the centers are stable for an indefinite period, while as the temperature increases the lifetime of these states decreases. At low temperatures the I_2^- molecule ions along one of the <110> directions can be reduced by irradiating the crystal with the polarized ultraviolet light along that <110> direction. As the temperature is increased this established orientation decreases. The temperature at which the disorientation rate is a maximum is called the disorientation temperature. In KI crystals the maximum rate of disorientation occurred at about 93°K. Above this temperature, the lifetime of the I_2^- center was too short for further measurements to be made.[45] Since a disorientation process of X_2^- centers was observed experimentally, the self-trapped hole must be capable of migrating in the crystal.

The X_2^- center may explain some of the luminescence properties of the alkali-halide crystals. The density of X_2^- molecule ions produced in the crystal would be expected to increase as the ionization density in the crystal increases simply because the density of ionized atoms increases. The ratio of electron traps due to impurities such as thallium to I_2^- molecules (which are assumed to be electron traps) would then decrease as the ionization density increases. The decay of the center formed by the capture of an electron by an I_2^- center, $I_2^- + e$, is here suggested as being responsible for the 3300A emission band in CsI

crystals. The lifetime of the I_2^- center only needs to be long enough to capture an electron in order for the 3300A band to result. No distinction is made here between this I_2^- + e center and the center proposed by Van Sciver as being an exciton captured by polarization of the crystal lattice.[11] Van Sciver suggested that the ultraviolet emission band of pure NaI resulted from the decay of this trapped exciton. The I_2^- molecule ion provides an explanation of the fact that the 3300A emission band is very weak for gamma-ray excitation of crystals which have electron traps such as thallium. The low ionization density produced by gamma rays would minimize the competition of I_2^- molecule ions with thallium centers for the capture of electrons. This concept treats the initial excitation of the thallium center as being by electron capture. The hole which is initially self-trapped can be thermally excited to the valence band and can migrate to the thallium center in a time which is characteristic of the temperature and the depth of the hole trap. For irradiation of the crystal by gamma rays then, the main process of exciting the thallium center would be by the successive capture of an electron and a hole. As the ionization density in the crystal increases and the density of I_2^- centers increases, other mechanisms of exciting the thallium center may occur. For cases in which the emission spectrum of the ultraviolet emission band of the pure crystal overlaps the absorption spectrum of the thallium center, sensitized luminescence may occur as well as optical excitation of the thallium center. The possibility of exciting the thallium center by the transport of excitons is not excluded. The fact that there is an overlapping of an emission band characteristic of

the pure CsI crystal with an absorption band in CsI(Tℓ) crystals due to the presence of thallium complicates the analysis of experimental data on the luminescence properties of CsI(Tℓ). A similar situation has been observed in NaI(Tℓ), where the ultraviolet emission band characteristic of the pure crystal overlaps a thallium absorption band.[31]

Some experiments performed by Teegarden and Weeks on the luminescence of pure KI crystals demonstrated a phenomenon which may be associated with the formation of I_2^- centers.[46] In these experiments it was shown that at the temperature of liquid nitrogen, KI crystals which were irradiated with F band light, after or during excitation by light in the region of the fundamental absorption band, emitted light in a band centered at about 3700A. This 3700A band corresponds to the 3300A band in CsI crystals. If the crystal was irradiated at liquid nitrogen temperature with photons having an energy of about 6.1 ev and subsequently irradiated with F-band light, a pulse of light from the 3700A band was observed. When the crystal was irradiated at room temperature with light in the fundamental absorption bands and then cooled to liquid nitrogen temperature, no emission was observed when the crystal was irradiated with F-band light. The existence of F centers in the latter case was demonstrated by photoconductivity experiments. The absence of the 3700A band for the case in which the F centers were produced at room temperature may be explained by the instability of I_2^- centers at this temperature.

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[46]K. Teegarden and R. Weeks, J. Phys. Chem. Solids 10, 211 (1959).

The emission of the 3700A band light can be explained by the capture at I_2^- centers of electrons freed from F centers. Teegarden and Weeks performed a thermoluminescence experiment using KI crystals which had been irradiated at the temperature of liquid nitrogen with light in the fundamental absorption bands.[46] There was a thermoluminescence peak at about 118°K; the emitted light was in the red region of the spectrum. The relative quantum yield of the 3700A emission band for F-band excitation was observed to decrease rapidly for temperatures above 100°K. This temperature corresponds very closely with the temperature of 93°K at which the disorientation rate of I_2^- molecules was observed to be a maximum.[45] Teegarden and Weeks attributed the thermoluminescence peak to a release of trapped holes as the temperature increases.[46]

Since the addition of Tl^+ ions to KI results in an increased production of I_2^- molecules, presumably by the Tl^+ ion capturing an electron during the X ray irradiations, thermoluminescence experiments in KI(Tl), or other thallium-activated alkali iodides, could be expected to give rise to thallium-activator luminescence. In fact, Van Sciver observed that the thermoluminescence emission spectrum of NaI(Tl) was very similar to the emission spectrum of the thallium band in NaI(Tl).[11] The ultraviolet emission band (about 3000A) was not observed in the thermoluminescence spectrum of any of the NaI(Tl) crystals investigated by Van Sciver.[11] The thallium content of one of the crystals was sufficiently low that light in the 3000A emission band could have been detected if it were present. The appearance of the thallium-emission band in the thermoluminescence experiments on NaI(Tl) is entirely in

accord with the concept of the recombination of a hole (as the temperature increases) with the electron trapped at the thallium-luminescence center.

Van Sciver observed that in NaI(Tℓ) the intensity of the thermoluminescence emission per unit energy deposited in the crystal was greater for gamma-ray excitation of the crystals than for alpha-particle excitation of the crystal. If the I_2^- centers compete with thallium centers and other electron traps in the crystal which are due to impurities (or crystalline defects) for the capture of an electron, then as the ionization density increases the I_2^- center may be expected to capture a larger fraction of the electrons produced by the exciting radiation. Thus, relatively more thermoluminescence emission would be expected for excitation by gamma rays than for alpha particles. Van Sciver suggested (1955 work) that relatively more "excitons" were produced as the ionization density increased and that this explained the difference in the production of thermoluminescence by gamma rays and alpha particles.[11]

In later experiments on the luminescence properties of NaI(Tℓ), Van Sciver (1960 work) attributed the ultraviolet emission band (2950A) characteristic of pure NaI to a crystalline defect.[31] One aspect of the experiments performed by Van Sciver (1960 work) is very suggestive that the $I_2^- + e$ center is responsible for the 2950A emission band in NaI. Measurements of the luminescence yield of the 2950A emission band in NaI showed that the yield increased as the intensity of irradiation increased for excitation with photons having an energy of 7.7 ev. For excitation with photons having an energy of 5.7 ev the luminescence yield of the

2950A emission band was independent of the incident photon intensity. It is here suggested that for excitation with 7.7-ev photons I_2^- centers and free electrons were formed; the band-gap energy in NaI is about 5.8 ev.[47] As the intensity of excitation increased, the equilibrium density of I_2^- molecule ions increased while the density of the other electron traps (associated with impurities) remained constant; this situation would lead to an increase in the yield of the 2950A emission band as the intensity of the excitation increases. For excitation with photons having an energy which is less than the band gap, it seems reasonable to assume that I_2^- centers are not formed with a high efficiency because of the small probability that the excited electrons will reach the conduction band. In this case the quantum efficiency of the 2905A band is expected to be constant (as observed) since the excited state is created directly by photon absorption rather than by recombination.

The X_2^- molecule ion seems to provide a reasonable explanation of some of the luminescence properties of the alkali halide crystals. The analysis of the experimental data on the luminescence properties of CsI(Tℓ) and NaI(Tℓ) are complicated by the overlapping of an emission band characteristic of the pure material with an absorption band in these crystals due to the presence of thallium. Since KI(Tℓ) crystals do not have such an overlapping of emission and absorption bands, these crystals may provide valuable information on the relative yield of the ultraviolet and the thallium-activator emission bands.

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[47]J. E. Eby, K. J. Teegarden, and D. B. Dutton, Phys. Rev. 116, 1099 (1959).

In conclusion, the ideas presented in this work have emphasized the role of the I_2^- center as a competitor with thallium centers for the capture of electrons in the crystal. As the ionization density increases the density of I_2^- centers increases, and at sufficiently large values of dE/dx the capture of electrons at I_2^- centers would be expected to predominate. This competition is very similar to that described by the Birks' model of the scintillation process, since the density of I_2^- centers increases linearly as the ionization density increases. As stated previously a competing process of this type leads to a dependence of the curve dL/dE versus dE/dx on the thallium concentration of the crystal in a manner similar to that predicted by the scintillation model. It appears, therefore, that the decreasing dL/dE at large dE/dx results from a process which is an intrinsic property of the ionization density in the crystal. This latter interpretation is in fact suggested by the experiments of Blue and Liu on the scintillation response of nonactivated alkali iodide crystals at 77°K.[48] Their experimental results cannot be interpreted, however, within the context of the present work by virtue of the very short integrating time constant used (2 microseconds) and the absence of a knowledge of the emission bands which contributed to the measured pulse heights. An understanding of the shape of the curve dL/dE versus dE/dx, which is nearly independent of activator concentration must, therefore, await further investigation.

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[48]J. W. Blue and D. C. Liu, IRE Trans. on Nuclear Sci. NS-9, 48 (1962).

CHAPTER VIII

SUMMARY

The goal of this experimental program was to provide experimental data on the scintillation response of CsI(Tℓ) crystals to monoenergetic charged particles in order to investigate some of the features of the scintillation model. In order to insure that the results obtained in this program would provide a critical test of the scintillation model, special attention was paid to technical effects which could influence the interpretation of the experimental data. For example, the effect of the pulse-analysis time on the relative scintillation response of CsI(Tℓ) to various charged particles was investigated. In addition, the emission spectra of the CsI(Tℓ) crystals were measured for excitation of the crystal by charged particles in order to correct the experimental data for the spectral sensitivity of the photomultiplier tube. The objectives of this program have been accomplished and the results obtained do provide a critical test of the scintillation model. The results have shown that dL/dE is a continuous function of dE/dx, as assumed in the scintillation model, within the accuracy of the experiments, when the effect of delta rays are considered. This leads to the conclusion that the light output of CsI(Tℓ) crystals is, in general, a nonlinear function of the energy of the exciting particle because dE/dx is a continuously varying function of the energy of the particle.

The shape of the dL/dE versus dE/dx curve for CsI(Tℓ) crystals has

been shown in this program to be nearly independent of the thallium content of the crystal. This is in distinct contrast to the predictions of the scintillation model.

An examination of the emission spectra of CsI(Tℓ) crystals has shown that light is emitted in an emission band, centered at about 3300A, characteristic of the emission band of the pure crystal when the crystal is excited by charged particles. The relative intensity of this luminescence with respect to the thallium-activator luminescence increases as the ionization density in the crystal increases and increases as the thallium content decreases. It has been suggested in this work that the emission band characteristic of pure CsI crystals is associated with the decay of an iodine molecule ion of the type I_2^- which has trapped an electron. Molecular ions of the type X_2^- have been identified in alkali halide crystals at low temperatures by investigations of the spin resonance and the optical spectra of these crystals. In these crystals the rate of production of molecular ions by X ray irradiation of the crystal can be increased by orders of magnitude by adding an electron trap, such as $Tℓ^+$ ions, to the crystal. It is thus evident from previous work that I_2^- centers are formed in thallium-activated alkali halide crystals when the crystals are subjected to ionizing radiation, and that the I_2^- centers compete with the thallium centers for the capture of electrons. On this basis it has been suggested here that thallium-activator luminescence results from the successive capture of an electron and a hole at a thallium center. Other means of exciting the thallium center have not been excluded.

The 3300A emission band of CsI overlaps the optical-excitation spectrum of CsI(Tℓ) for thallium-activator luminescence. Thus some thallium centers in CsI(Tℓ) are excited by optical emission in the 3300A band. The overlapping of an emission band characteristic of the pure CsI crystal with an absorption band due to the presence of thallium in CsI(Tℓ) complicates the interpretation of experimental data on the luminescence properties of this crystal. This same situation exists in NaI(Tℓ). It has been suggested that an investigation of the luminescence properties of KI(Tℓ) would be worthwhile because the ultraviolet emission band of KI (about 3700A) does not overlap an absorption band in KI(Tℓ) due to the presence of thallium.

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