Scintillation Process in CsI(Tl). I. Comparison with Activator Saturation Model*

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The scintillation efficiency of CsI(Tl) crystals, containing varying amounts of Tl, has been measured upon excitation by gamma rays, protons, and alpha particles in the keV and MeV region. The purpose of this program was to provide a critical test of a previously proposed phenomenological model of the scintillation process in activated alkali halides. In order to achieve this goal, special attention has been paid to technical effects which might distort the data, viz., effects dependent on the emission spectrum and the pulse analysis time. It is concluded that the variation of the scintillation efficiency as a function of the stopping power of the incident particle is predicted by the scintillation model when the effect of secondary electrons is taken into account. It is found that the shape of the scintillation efficiency versus dE/dx curve is very nearly independent of Tl concentration. It is concluded from this fact that the decline in scintillation efficiency for highly ionizing particles is not due to saturation of activator centers.

I. INTRODUCTION

IT is well known that the scintillation efficiency of activated alkali iodides is less for highly ionizing particles (alphas, C^{12} ions, etc.) than for protons. (Scintillation efficiency is here defined as dL/dE, the slope of a pulse height versus energy curve.) This decrease in scintillation efficiency has often been attributed to "saturation of the luminescence centers" for a particle of high specific-energy loss, dE/dx. In order to investigate the validity of the saturation concept, a phenomenological model of the scintillation process was proposed by Murray and Meyer[1] and the consequences of this model were examined theoretically. It was concluded that the saturation process could reasonably account for the observed shape of the scintillation efficiency versus dE/dx curve for a crystal of given Tl content. This model further predicted that the dL/dE versus dE/dx curve would depend on the Tl concentration. Although there is a wealth of experimental information on the pulse-height response of activated alkali iodides to various charged particles, the detailed experimental results necessary to check the predictions of the scintillation model have not been available. The purpose of the present work is to examine in detail the scintillation efficiency of a family of CsI(Tl) crystals, having different Tl contents, in order to provide a critical test of the assumptions and consequences of the proposed scintillation model. In particular, we wish to examine the following aspects of this model: (a) the assumption that dL/dE is a continuous function of dE/dx for a given particle and that the discontinuity in dL/dE at a given dE/dx for different particles depends upon the production of secondary electrons by the exciting particle; (b) the prediction that dL/dE is nowhere strictly a constant in dE/dx but is continuously varying; (c) the prediction that dL/dE as a function of E should pass through a minimum in that energy region where dE/dx is a maximum; and (d) the prediction that the shape of dL/dE versus dE/dx depends on Tl concentration.

In order to achieve these objectives it is necessary to pay attention to technical effects which might seriously distort the results. There are two technical effects which must be particularly considered in the present work. First, the scintillation model treats only light emitted from activator centers. It is thus necessary in these experiments that the system respond only to light characteristic of the activator center. Changes in the emission spectrum associated with the identity or stopping power of the charged particle must be taken into account. Second, the scintillation model describes only the *total* number of photons emitted, integrated over all time. It is thus necessary that scintillation pulses be integrated for a period of time which is long compared to the characteristic decay time of the pulse. Changes in the decay time associated with the identity or stopping power of the crystal for the charged particle must be taken into account.

The present experimental program was carried out with crystals of CsI(Tl). The experiments consisted of measurements of the scintillation pulse height as a function of energy for monoenergetic gammas, protons, and alphas in several crystals of CsI(Tl) whose Tl content ranged from 0.002 to 0.31 mole %. In auxiliary experiments, the emission spectra of these crystals were measured upon excitation by the same radiations. These results yield dL/dE versus dE/dx over a wide range of dE/dx, from which it is concluded that items (a), (b), and (c) above are found to be in agreement with the scintillation model, whereas item (d) is in sharp contrast to the model. It is concluded that the decline in dL/dE at large dE/dx is *not* due to saturation of the luminescence centers.

Discussion of experimental techniques, data analysis, etc., in the following sections is condensed. A detailed description is available elsewhere.[2]

* This work was included in a thesis submitted by R. Gwin in partial fulfillment of the requirements for the degree of Doctor of Philosophy at the University of Tennessee.
† Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.
[1] R. B. Murray and A. Meyer, Phys. Rev. **122**, 815 (1961).

[2] R. Gwin and R. B. Murray, Oak Ridge National Laboratory Report ORNL-3354, 1962 (unpublished).

II. EXPERIMENTAL METHODS

The CsI(Tl) crystals used in this experimental program were grown by the Stockbarger method. Two of the crystals were grown in an available furnace at this laboratory, while all other crystals were obtained from the Harshaw Chemical Company. Spectrographic analysis of all crystals showed that the cesium iodide contained small amounts of other alkali metals: The concentration of potassium was about 0.02% by weight, while other alkali metals were present in lower concentrations. There were traces of other elements; silver, magnesium, iron, and silicon were present in concentrations of order 5 parts per million (ppm).

Crystals used in these experiments were cut from the various ingots to have a diameter of 2.5 cm and a thickness of 0.2 cm. The Tl content of each crystal was determined by polarographic analysis of a sample taken from the ingot immediately adjacent to the slice used as a scintillation crystal. Each time the photomultiplier system was assembled both surfaces of the crystal were polished to a smooth, clear surface. Light from the scintillation pulse was detected by an RCA C-7261 photomultiplier (now available as the RCA 7326) whose photocathode has an S-20 response. The crystal was optically coupled to the photomultiplier through a cylindrical Lucite light pipe.

Current pulses from the photomultiplier were integrated at the tube anode by an *RC* circuit whose time constant was $\sim$250 μsec. Amplified voltage pulses were analyzed by a multichannel pulse-height analyzer. Two separate amplifying and pulse-analyzing systems were used in these experiments, one for a pulse-analysis time of 1 μsec and the other for a pulse-analysis time of 7 μsec. "Pulse-analysis time" refers to the time following initiation of the pulse at which the pulse amplitude is measured. The 7-μsec system consisted of a DD-2 amplifier modified by the addition of 7-μsec delay lines, and an Atomic Instrument Company 20-channel analyzer which was equipped with a long-pulse adapter. The 1-μsec system was composed of an A-8 amplifier and a Nuclear Data ND-120 Analyzer. The pulse-analysis system was calibrated in the region of interest after recording each pulse-height spectrum by the use of a precision pulse generator whose output was monitored with a Rubicon laboratory potentiometer. All pulse-height measurements were thus referred to the potentiometer. With this method pulse-height measurements do not depend on the linearity or long-term stability of the electronic system or a knowledge of the analyzer zero.

The basic experimental method consisted of the measurement of the light output of a particular 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 pulse height per unit energy L/E_γ for this standard was arbitrarily assigned the value 1.00. Values of L/E for other particles are thus normalized to L/E for 662-keV gammas. In all measurements the pulse height L was taken to be that voltage corresponding to the mean of the full-energy peak in the pulse-height spectrum. For most incident particles the full-energy peak was symmetrical and the mean was determined as the midpoint of the distribution at $\frac{2}{3}$ maximum. In some cases (low-energy gamma rays) the peak was asymmetric; corrections were applied to account for the asymmetry as discussed later. Care was taken to insure that the results were not influenced by a dependence of photomultiplier gain on count rate. All experiments were performed at room temperature.

III. EXPERIMENTAL RESULTS

A. Scintillation Response to Gamma Rays

The response of the various CsI(Tl) crystals was measured as a function of gamma-ray energy in the interval 10.7 keV to 2.75 MeV. 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 *K* capture. Although these sources do not emit monoenergetic radiation, they can be used without introducing large uncertainties in the experimental results. Assuming that each component of a *K*-shell x-ray source produces a Gaussian pulse-height distribution, and assuming that the pulse height is proportional to energy over the spread of *K* x-ray energies, a short calculation[2] shows that the peak of the distribution occurs at a pulse height corresponding to an average energy $\bar{E}$ which depends on instrumental line width and the number N_i of x rays of energy E_i. For the broad resolution characteristic of the kev region $\bar{E}$ approaches the "source average energy" $E_s = \sum N_i E_i / \sum N_i$. In the present work $\bar{E}$ was calculated for all *K*-shell x-ray sources. The maximum departure of $\bar{E}$ from E_s was 1%. Values of the energies and relative intensities of the *K*-shell x-ray components were taken from the literature,[3] as were the energies of nuclear gamma rays.[4]

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 incident gamma, giving rise to the "*K*-escape peak," and (2) statistics of the photomultiplier response for weak light pulses. In connection with item (1), an estimate of the effect of the *K*-escape peak was made on the basis of the work by Axel[5] and knowledge of the instrumental linewidth for the various crystals used. The largest correction made for the effect of the *K*-escape peak was about 2.5%. With regard to item (2) the asymmetry of the peak is determined by a param-

[3] G. J. Nijgh, A. H. Wapstra, and R. Van Lieshout, in *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

[4] D. Stominger, J. W. Holland, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).

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

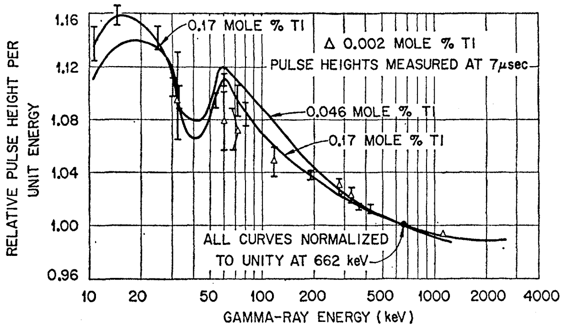

FIG. 1. Measured pulse height per unit energy for gamma rays on CsI(Tl) crystals of varying Tl content for a pulse-analysis time of 7 μsec. Curves obtained with 1-μsec analysis are very similar.

eter δ which depends on the statistics of electron multiplication in the dynodes. The value of δ used for the correction of data in the present experiments was 1.3±0.6, which represents an average of the values quoted by Wright[6] and, in a recent study, by Prescott.[7] In order to make the correction it was further necessary to know the number of photoelectrons ejected in a low-energy scintillation event. This information was obtained from auxiliary experiments in which the current from the photocathode was measured during steady-state excitation of the crystal by a known flux of monoenergetic alpha particles. The largest correction from this effect amounted to 6.8%.

Experimental data from the gamma-ray experiments are shown in Fig. 1, in which pulse height per unit energy, L/E, is plotted as a function of gamma-ray energy. All curves are normalized to unity at 662 keV. Figure 1 shows the data at 7-μsec analysis time only; data obtained at 1 μsec are very similar. Error limits are shown for a few points on the curves and include uncertainties in reproducibility of the individual points, corrections for asymmetry, and knowledge of the source energy. The same type of behavior shown in Fig. 1 has been previously observed in NaI(Tl).[8,9] It is seen in Fig. 1 that the shapes of the curves are relatively insensitive to Tl concentration.

As indicated previously, data of Fig. 1 are normalized at 662 keV for all three curves. The actual light intensity (pulse height) for a particular gamma ray is of course a function of Tl concentration. This function was measured using all the available crystals, upon excitation by the 662-keV gamma, and is shown in Fig. 2. Data of Fig. 2 were measured with a 7-μsec analysis time; analysis at 1 μsec results in a very similar curve which is not shown. There is no evidence in Fig. 2 (or in the data obtained at 1 μsec) of a decline in the pulse height at large Tl concentrations associated with concentration quenching. The data of Tsirlin[10] on the shape of the pulse height versus Tl concentration for CsI(Tl) crystals are in contrast with the data presented in this paper.

B. 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 ORNL 5.5 MV Van de Graaff generator. The analyzed proton beam was scattered from a thick tantalum foil; protons scattered into a particular solid angle entered the entrance slit of a 60 deg charged-particle analyzing magnet. The CsI(Tl) crystal assembly was located at the exit slit of this magnet. The analyzing magnet was carefully calibrated, prior to the scintillation experiments, by standard techniques. All calibration measurements were referred to the $Li^7(p,n)Be^7$ threshold which was taken to be 1880.7 keV.[11] Auxiliary experiments were performed to determine whether a measurable difference existed between the optical collection efficiency in the scintillation counter for proton events versus gamma-ray events. No difference was observed within the limits of uncertainty of the experiment, which amounted to about ½%. The scintillation crystal was mounted in a manner which was designed to minimize the difference in the optical collection efficiency between gamma-ray excitation and charged-particle excitation of the crystal (see Ref. 2).

Experimental results for the 0.046 mole % Tl crystal are shown in Fig. 3. This figure shows the data obtained at both pulse-analysis times. All data points are normal-

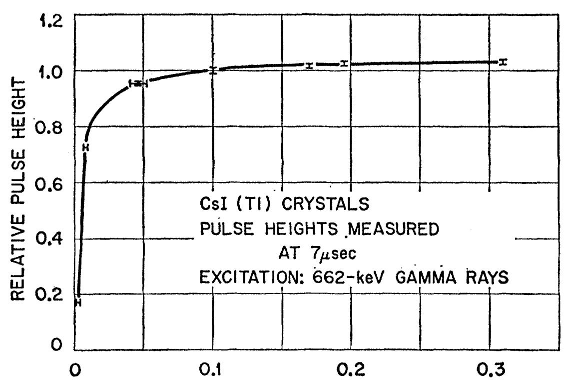

FIG. 2. Relative pulse height per unit energy resulting from the excitation of CsI(Tl) crystals with 662-keV gamma rays as a function of Tl content of the crystal. The voltage pulse was measured at 7 μsec. The curve obtained with 1-μsec analysis time is very similar. Horizontal error bars are estimates of the uncertainty in Tl concentration; vertical error bars are based on reproducibility of the individual points.

[6] G. T. Wright, J. Sci. Instr. **31**, 462 (1954).
[7] J. R. Prescott, University of Alberta (private communication).
[8] W. C. Kaiser, S. I. Baker, A. J. MacKay, and I. S. Sherman, IRE Trans. Nucl. Sci. **9**, 22 (1962); also see references therein.
[9] W. W. Managan, IRE Trans. Nucl. Sci. **9**, 1 (1962).
[10] Yu. A. Tsirlin, S. N. Komnik, and L. M. Soifer, Opt. i Spektroskopiya **6**, 422 (1959) [translation: Opt. Spectry. (USSR) **6**, 265 (1959)].
[11] J. Marion, Rev. Mod. Phys. **33**, 139 (1961).

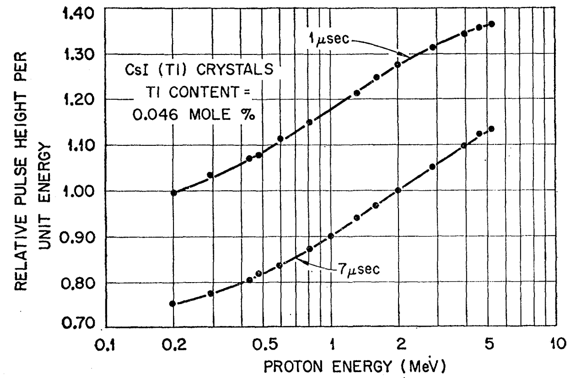

FIG. 3. Relative pulse height per unit energy from proton excitation of a CsI(Tl) crystal of 0.046 mole % Tl. All values are normalized to $(L/E)_\gamma=1$ for 662-keV gammas, measured at the same analysis time.

ized to $L/E=1$ for 662-keV gammas measured at the same pulse-analysis time; i.e., the 7-μsec curve was measured relative to the gamma-ray standard at 7 μsec, and the 1-μsec curve was measured relative to the gamma-ray standard at 1 μsec. Presentation of the two curves on the same drawing does not imply that the pulse height at 1 μsec is greater than that at 7 μsec; in fact, the converse is true.

The data of Fig. 3 show that for a pulse-analysis time of 1 μsec L/E for protons at 662 keV is greater than L/E for gammas at 662 keV, while at 7 μsec the converse is true. During the course of 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 1 μsec the proton pulse was greater than the gamma-ray pulse, while at 7 μsec the gamma-ray pulse was greater than the proton pulse. This is in accord with detailed measurements of the time dependence of scintillation pulses from CsI(Tl) excited by particles producing different ionization densities in the crystal.[12] These observations demonstrate that the detailed nature of the relative scintillation response of CsI(Tl) is a complex function of pulse-analysis time and ionization density in the crystal.

C. Scintillation Response to Alpha Particles

The scintillation response of CsI(Tl) crystals to alpha particles was measured in the energy range from 58 keV to 10 MeV. The experimental methods were the same as in the proton experiments. Alpha-particle energies above 5 MeV were obtained by accelerating doubly charged helium ions. Experimental results from the crystal having a Tl content of 0.046 mole % are shown in Fig. 4. Again, both curves are normalized to $L/E=1$ for 662-keV gammas measured at the same analysis time.

[12] R. S. Storey, W. Jack, and A. Ward, Proc. Phys. Soc. (London) **72**, 1 (1958).

The minimum in each curve of Fig. 4 occurs at the energy (∼1 MeV) corresponding to the maximum specific energy loss for alpha particles. This feature is predicted by the scintillation model (see Ref. 1, Fig. 8) as a consequence of the assumption that dL/dE is a continuous and monotonically decreasing function of dE/dx. Since dE/dx as a function of energy passes through a maximum, it follows that dL/dE as a function of energy must pass through a minimum. The minimum in L/E of Fig. 4 produces a kink in the pulse height versus energy curve, see Fig. 5. The increasing slope at large energies, observed in the lower half of Fig. 5, has been frequently reported and is a well-known characteristic of heavy particles. The kink below ∼1 MeV, to the authors' knowledge, has not been observed previously.

IV. DATA ANALYSIS

The data of Fig. 1 represent the response to gamma rays and not to monoenergetic electrons. The response function to electrons is not the same as that to gammas, since a monoenergetic gamma ray of energy E does not simply produce an electron energy E in the crystal. It is possible, however, to derive the electron response function from the data of Fig. 1 following a procedure outlined previously[13] for the case of NaI(Tl). The detailed calculation is available elsewhere.[2] Briefly stated, it is assumed that for incident gamma rays of energy less than 80 keV, only photoelectric interactions take place. Vacancies in the K-, L-, and M-shell levels of Cs and I, resulting from a photoelectric event, are treated by a simple cascade model which leads to the electron response function below 80 keV. At higher energies Compton and pair-production processes contribute (or dominate) and the electron response function is derived on the basis of a previously developed Monte Carlo calculation.[13] In calculating the electron response function, no distinction was made between the three curves of Fig. 1, as they are so nearly the same. The

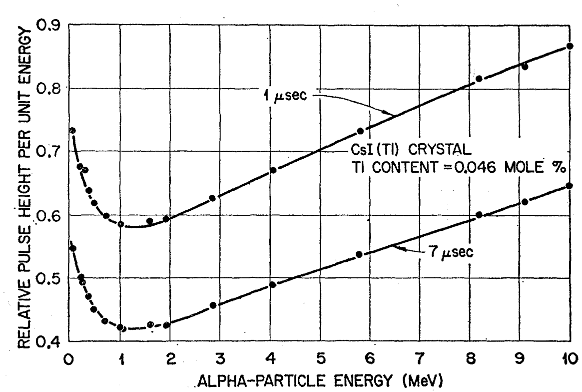

FIG. 4. Measured values of pulse height per unit energy for alpha particle excitation of the CsI(Tl) crystal of 0.046 mole % Tl. All values are normalized to $(L/E)_\gamma=1$ for 662-keV gammas, measured at the same analysis time.

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

calculation was performed with data from the 0.046 mole percent crystal. The electron response function, derived from the data of Fig. 1 as outlined above, is shown in Fig. 6. This function is based on a 7-μsec analysis time. The ordinate of Fig. 6 is still normalized to a gamma-ray response of unity at 662 keV. The electron response function based on 1-μsec analysis is very similar and is not shown. Uncertainty limits shown in Fig. 6 are based on experimental uncertainties in the gamma-ray measurements and on estimates of the uncertainties introduced by the model used to describe gamma-ray interactions in the crystal. Uncertainties below ~10 keV are large, as indicated by the dashed curve in Fig. 6.

It is of interest to note that the pulse height per unit energy for electrons passes through a maximum at low energies. This maximum in the electron response function is responsible for the dip near 40 keV in the gamma response curves of Fig. 1. This feature arises from the details of the photoelectric process and has been described elsewhere.[14]

Having obtained the pulse height per unit energy for electrons, protons, and alpha particles, it is now necessary to analyze these data to obtain the scintillation efficiency dL/dE as a function of dE/dx. As indicated earlier, it is first necessary to examine effects dependent on the emission spectrum and the pulse decay time. Measurements of the emission spectra of the various crystals, upon excitation by electrons, protons, and alphas, are described in an accompanying paper.[15] Knowing the emission spectra and the approximate photomultiplier response function, it is possible to determine how the observed data are influenced by changes in the spectrum. The observed pulse height is proportional to the integral

$$\int_0^\infty I(\lambda)P(\lambda)T(\lambda)d\lambda,$$

where $I(\lambda)$ is the (normalized) emission spectrum of the crystal, $P(\lambda)$ is the photocathode sensitivity function, and $T(\lambda)$ is a transmission function, characteristic of the crystal geometry and reflector optics, which should be close to unity for all λ over the emission spectrum. For present purposes it is assumed that the observed emission spectrum represents the product $I(\lambda)T(\lambda)$. Taking $P(\lambda)$ as a typical S-20 response function, the above integral was evaluated for excitation of the crystals by electrons, protons, and alphas, and it was found that these integrals were constant within the uncertainty of the spectral measurements, i.e., about 3%. Since the integrals were found to be nearly constant for the nominal S-20 response function, it was not necessary to measure $P(\lambda)$ for the particular photo-

[14] R. Gwin and R. B. Murray, IRE Trans. Nucl. Sci. **9**, 28 (1962).

[15] R. Gwin and R. B. Murray, following paper, Phys. Rev. **131**, 508 (1963).

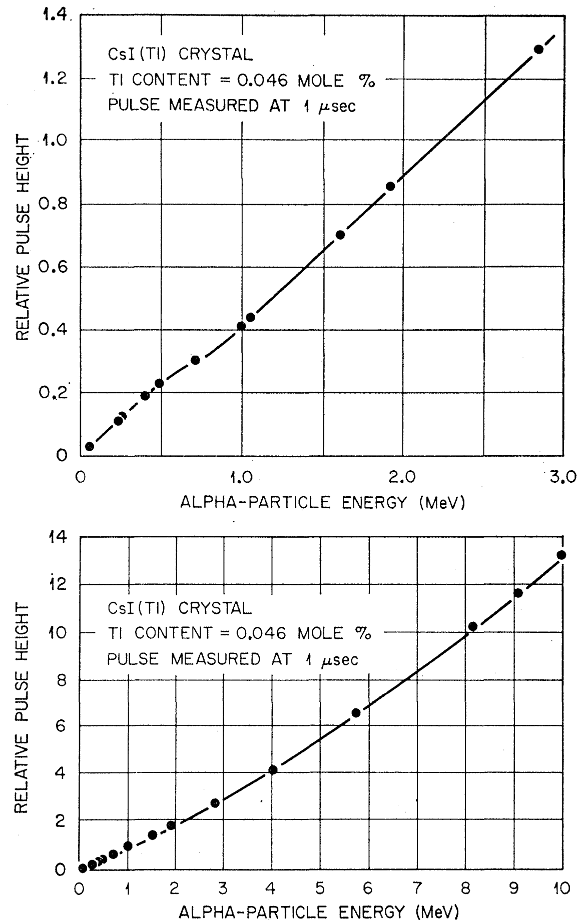

FIG. 5. Relative pulse height as a function of energy for alpha-particle excitation, from Fig. 4. Pulse was measured at 1 μsec. The scales of the ordinates are arbitrary and are different in the two figures.

multiplier used. The final result is that the pulse heights observed in the scintillation response experiments can be used, without correction, as a direct measure of the light output characteristic of the Tl luminescence. This feature arises from the fact that changes in the emission spectrum occur principally at short wavelength, where $P(\lambda)$ is small, and do not sensibly affect the integral.

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.[12] The relative values of the measured pulse heights for particles of different dE/dx thus depend upon the pulse-analysis time. In the present program measurements were made for pulse-analysis times of 1 and 7 μsec in order to determine whether there was a gross dependence of the scintillation efficiency on pulse-analysis time. It is concluded that the qualitative behavior is the same at both analysis times. The experimental data of Storey, Jack, and Ward[12] show that about 80% of the light in a scintillation pulse from CsI(Tl) produced by gamma rays is emitted in the first 7 μsec (assuming that all the light is contained within two exponentially decaying components, one of lifetime $\sim\frac{1}{2}$ μsec and one of lifetime ~ 7 μsec). For alpha-particle excitation the corresponding quantity is 90%. Thus, for present purposes data obtained at a pulse-analysis time of 7 μsec permit a reasonable comparison with the scintillation model. A more meaningful comparison can be made by correcting the 7-μsec data to "infinite analysis time," i.e., to the total light output contained within the two components. This correction is possible on the basis of previous data[12] and has been applied in the analysis. The scintillation efficiency curves so calculated, and presented in Sec. V, are very similar to the curves based directly on the 7-μsec analysis time. It should be understood that *only* the fast scintillation components are being considered. No attempt is made to account for the contribution of possible long-lived phosphorescence.

In order to compare the results of the present experiments directly with the scintillation model, the slope dL/dE of the pulse-height versus energy curve must be obtained. The function dL/dE was determined from the relation,

$$dL/dE \equiv (L/E) + E(d/dE)(L/E). \quad (1)$$

This proves to be a convenient prescription for analysis of the functions in the present work, since L is nearly proportional to E and $dL/dE \approx L/E$ as a first approximation. The second term on the right side of Eq. (1), which must be obtained by graphical analysis, makes a minor contribution to dL/dE. Errors introduced in the graphical analysis are, therefore, minimized.

Finally, it is necessary to know the specific energy loss of the various particles in cesium iodide as a function of their energy. For protons and alpha particles, dE/dx as a function of energy was taken from a previous calculation.[1] Values of dE/dx for electrons were calculated by A. Meyer of this Laboratory using recent range-energy curves for electrons given by Kanter and

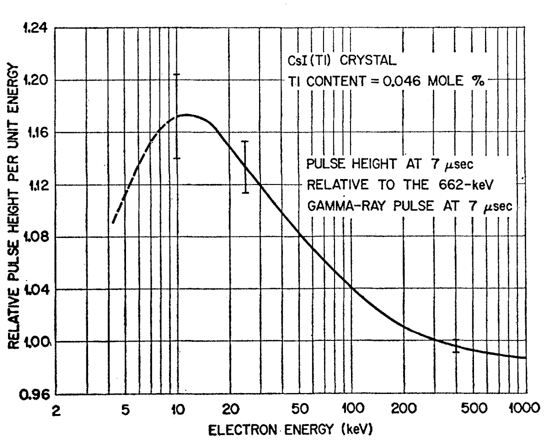

FIG. 6. Derived value of pulse height per unit energy for electron excitation of a CsI(Tl) crystal having a Tl content of 0.046 mole %. The dashed curve indicates large uncertainties.

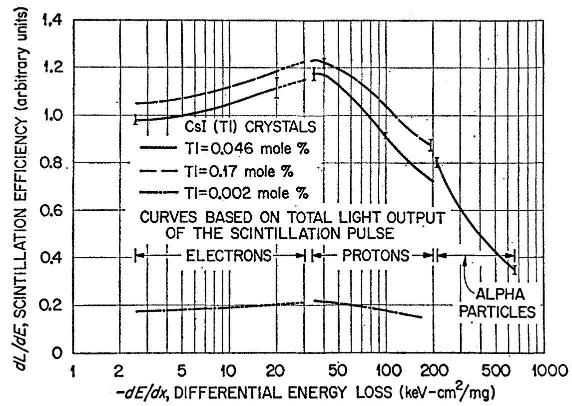

FIG. 7. Scintillation efficiency as a function of dE/dx for CsI(Tl) crystals of varying Tl content. Values of dL/dE are based on the total light contained within the two fastest components of the scintillation pulse as discussed in text. Error limits are based on uncertainties in measurements and data analysis.

Sternglass.[16] The calculated curve of dE/dx as a function of energy for electrons in cesium iodide is given elsewhere.[2]

V. RESULTS AND COMPARISON WITH MODEL

The results of the measurements and data analysis are presented in Fig. 7, in which the scintillation efficiency dL/dE is plotted as a function of dE/dx for three crystals of different Tl concentration. These curves are based on pulse heights extrapolated to include the total light emitted in the $\frac{1}{2}$- and 7-μsec components, as discussed in Sec. IV. It is seen in Fig. 7 that the scintillation efficiency is apparently a continuous function in passing from electrons to protons, within the uncertainties in the present experiments and data analysis. Uncertainties in dL/dE for low-energy electrons, shown dashed in Fig. 7, are not well known but should not exceed $\pm 10\%$. Although dE/dx values for electrons and protons do not completely overlap, it should be recognized that dE/dx for electrons is not well known in the high-dE/dx region. A large uncertainty in dE/dx, however, does not affect the conclusion since dL/dE for both electrons and protons is nearly flat in the overlap region.

In passing from protons to alpha particles, it is clear that the curves do not join smoothly, and that the discontinuity is greater than the combined uncertainties in the two curves. A discontinuity of the magnitude shown is anticipated, however, on the basis of a contribution to dL/dE of alpha particles from high-energy secondary electrons which escape the primary column of high ionization density and produce light in a virgin region of the crystal with a high scintillation efficiency. This effect has been treated previously[17] and, on the basis of that analysis, it is possible to obtain a rough estimate of the contribution of this effect. It is estimated

[16] H. Kanter and E. J. Sternglass, Phys. Rev. **126**, 620 (1962).
[17] Axel Meyer and R. B. Murray, Phys. Rev. **128**, 98 (1962).

that for 10-MeV alpha particles, about 7% of dL/dE is due to the contribution of energetic secondary electrons. This estimate is to be compared with the observed discontinuity of $\sim$10%; the uncertainty in dL/dE is about $\pm$2% for both the proton curve and the alpha-particle curve. It is concluded that the observed discontinuity is consistent with the estimated secondary-electron effect. On this basis the experimental results are consistent with the original assumption of the scintillation model that the scintillation efficiency is a continuous function of dE/dx, independent of the identity of the exciting particle. Secondary electrons would not be an important consideration in the case of primary excitation by electrons as shown in Ref. 17. In this context the term "scintillation efficiency" means the efficiency in the primary ionization column, and is contrasted with the measured total scintillation efficiency which includes the effect of secondary electrons. This distinction is discussed in detail elsewhere.[17]

A second conclusion to be drawn from Fig. 7 is that dL/dE is a continuously varying function; i.e., dL/dE is not constant over any significant range of dE/dx. Thus, L is not a strictly linear function of the energy, over an appreciable energy interval, for any of the particles available in these experiments. This conclusion must be tempered with the understanding that dL/dE is a slowly varying function of energy, especially for electrons and protons in the MeV region, so that for many purposes L can be considered as a linear function of energy to a good approximation.

The final point concerns the dependence of the shapes of the curves of Fig. 7 on activator concentration. On the basis of the scintillation model, the shape of the scintillation efficiency curve depends on Tl concentration as exhibited in the top half of Fig. 8. The three calculated curves have been normalized to the same magnitude at low dE/dx. It is seen that these curves predict significantly different values of dL/dE at high dE/dx. For comparison, the experimental curves of Fig. 7 have been normalized at low dE/dx and are shown in the lower half of Fig. 8. Additional data from a 0.31 mole percent Tl crystal are included. It is seen in Fig. 1 that the normalized experimental curves all have very nearly the same shape, in distinct contrast with the divergent curves predicted by the scintillation model. These experimental results (Fig. 8) are also in disagreement with the work of Tsirlin.[10] Tsirlin's data[10] show a distinct decline in the ratio of the pulse height produced by alpha particles ($\sim$5 MeV) to that produced by gamma rays as the Tl content of the crystal decreases. The results of the present work shows that the ratio of the pulse height produced by highly ionizing protons (295 keV) to that by gamma rays is essentially constant over the range of Tl concentrations investigated. It is concluded that the activator saturation mechanism described in the scintillation model is contradicted by experiment, and that this saturation mechanism is, therefore, not responsible for the decline in scintillation efficiency at large dE/dx.

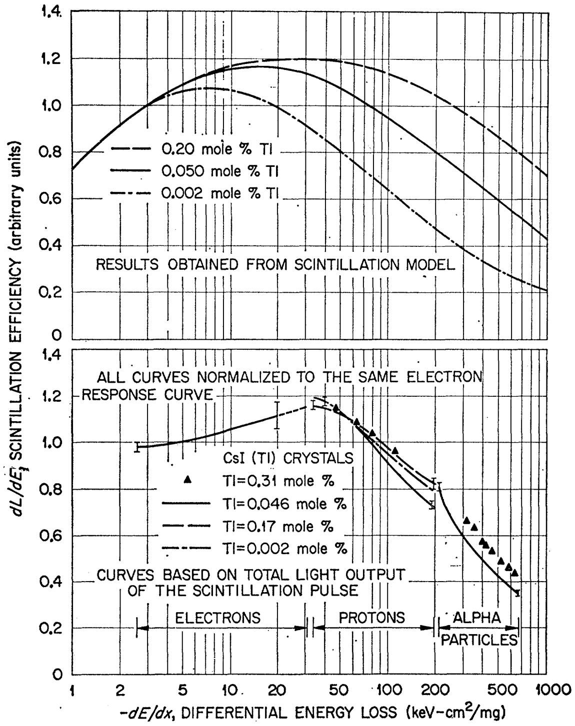

FIG. 8. (Upper) Calculated curves of scintillation efficiency versus dE/dx from saturation model of Ref. 1. All curves normalized at low dE/dx. (Lower) Experimental curves based on total light output in the two fastest components of scintillation pulse. All curves normalized at low dE/dx.

VI. CONCLUSION

In conclusion, the experimental results of this paper indicate that the activator saturation mechanism does not contribute significantly to the decline in scintillation efficiency at high specific energy loss. It appears instead that the decrease in 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.[18] 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 μsec) 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 the activator concentration must await further investigation.

It would be of considerable interest to examine separately the intensities of the Tl band and the ultraviolet emission band which is characteristic of the pure

[18] J. W. Blue and D. C. Liu, IRE Trans. Nucl. Sci. 9, 48 (1962).

material, as a function of ionization density and Tl content. Unfortunately, this is not possible in NaI(Tl) or CsI(Tl), as a consequence of the overlap between the ultraviolet band and the Tl absorption. This overlap does not occur in KI(Tl), however, so that the crystals of KI(Tl) would provide a favorable medium for such a study.

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Scintillation Process in CsI(Tl). II. Emission Spectra and the Possible Role of Self-Trapped Holes*

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The luminescence emission spectra of CsI(Tl) crystals containing varying amounts of Tl have been measured upon excitation by gamma rays, protons, and alphas. The principal features include a broad Tl emission band centered near 5500 Å and a narrower band centered near 3300 Å which is characteristic of the host crystal. This 3300-Å band is analogous to near-ultraviolet emission bands observed in NaI and KI. It is found for CsI(Tl) that the intensity of the 3300-Å band relative to the Tl band is greatest upon excitation by radiation of high dE/dx. It is suggested that the 3300-Å band arises from the recombination of free electrons with self-trapped holes. Evidence from other investigations supporting this hypothesis is summarized.

I. INTRODUCTION

In connection with the experimental program described in Part I,[1] it was necessary to measure the emission spectra of the various CsI(Tl) crystals used upon excitation by radiations of considerably different stopping power. The results of these measurements are of some intrinsic interest, and for this reason are presented here. These measurements do not constitute the first studies of the emission spectrum of CsI(Tl) crystals. In fact, several authors[2–5] have previously reported the emission spectrum, with results which are not in complete agreement. The present measurements, however, are unique in that they present a systematic study of the spectra upon changing both the Tl content and the average stopping power of the exciting particle.

II. EXPERIMENTAL METHOD AND RESULTS

Emission spectra were measured with a Bausch and Lomb 500-mm focal length grating monochromator which had a nominal dispersion of 33 Å per mm. Light from the exit slit of the monochromator was detected with an RCA-7265 photomultiplier. The relative spectral response of the monochromator-photomultiplier system was measured with a tungsten-filament lamp whose emission spectrum was known from a previous calibration at the National Bureau of Standards. Emission spectra were measured upon excitation by 50- and 250-keV x rays, 1.4- and 4.4-MeV protons, and 2.0- and 8.7-MeV alpha particles. The x-ray beam was filtered[6] to provide a reasonably limited band of x-ray energies: The lower energy beam extended from about 30 to 50 keV, and the higher energy beam extended from 150 to 275 keV. Monoenergetic protons and alphas used for excitation were obtained with the 5.5-MV Van de Graaff generator. The current density of both protons and alphas on the crystal surface was limited to a maximum value of 10^{-9} A/cm^2. The total light output of the crystal was continuously monitored to account for fluctuations in beam current.

Results of the spectral measurements are summarized in Figs. 1 through 3, in which the ordinate has been

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† Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

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