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## The Fluorescent Decay of CsI(Tl) for Particles of Different Ionization Density

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*Abstract.* New measurements of the decay of the fluorescence produced in CsI(Tl) have been made for different particles. The detailed shape of the decay is found to depend on the average ionization density  $\rho$ . The lifetimes  $\tau$  of the main components are  $0.425 \pm 0.025 \mu\text{sec}$  for 4.8 MeV alpha particles,  $0.52 \pm 0.01 \mu\text{sec}$  for 2.2 MeV protons,  $0.595 \pm 0.02 \mu\text{sec}$  for 8.6 MeV protons and  $0.70 \pm 0.025 \mu\text{sec}$  for 0.66 MeV electrons. The variation of  $\tau$  and  $\rho$  is very similar to that observed by others for the variation of  $\tau$  with temperature for alpha particles, which suggests that the former may be explained in terms of variations in the effective local temperature. It is shown that the differences in  $\tau$  are sufficient to allow a single crystal detector to be used in experiments where particle type as well as energy determination are required.

### § 1. INTRODUCTION

IN the scintillation counter, the light produced by an ionizing particle in a fluorescent material is converted into an electrical current pulse with the same time variation. It is generally believed that the rate of decay of the main pulse is independent of the type of particle, and that the scintillation counter is only an energy spectrometer. While it is possible to deduce indirectly the type of particle being counted, it would be desirable, from the point of view of achieving a high counting efficiency and simplicity of operation, if the shape of the pulse could be used to identify the particle.

It is therefore of practical, as well as theoretical, interest to consider whether there is any variation for different particles in  $\tau$ , the mean life of the main pulse in CsI(Tl), a commonly used scintillating material. (Although the fluorescent efficiency of CsI(Tl) is less than that of NaI(Tl), its excellent physical and mechanical properties make it preferable in most applications.)

Van Sciver and Hofstadter (1951), using an RCA 5819 photomultiplier, have found a value for  $\tau$  of  $1.1 \pm 0.1 \mu\text{sec}$  for fast electrons in CsI(Tl), whereas Bonanomi and Rossel (1952), using the same type of photomultiplier, give a value of  $0.6 \mu\text{sec}$  for polonium alpha particles. Knoepfel *et al.* (1956), using an RCA 6655, and also measuring  $\tau$  for alpha particles obtain a value of  $0.55 \mu\text{sec}$ . Possibly these results have not been interpreted as due to difference in particle, because it is known that other factors can significantly influence the measured lifetime.

Knoepfel *et al.* (1957) have found that  $\tau$  for alpha particles in CsI(Tl) varies with temperature. Eby and Jentschke (1954) have shown that  $\tau$  is the same for

alphas and electrons in NaI(Tl), but varies with thallium concentration. From these last results, it would be reasonable if the different values of  $\tau$  measured in CsI(Tl) were at least partly due to a difference in the concentration of thallium in the crystals used. Kallmann and Brucker (1957), using an RCA 5819, have measured  $\tau$  for fast electrons and alpha particles in a range of organic and inorganic crystals not including CsI(Tl) and find no significant difference.

Summarizing the results of all the above experiments, it can be stated that while different values of  $\tau$  have been observed for alpha particles and electrons in CsI(Tl), no such effect has been found in the main current pulse of other scintillators, and the measured values of  $\tau$  may be expected to vary with temperature, concentration of thallium and, possibly, type of photomultiplier.

In the experiments described below, these last three factors were kept constant, and  $\tau$  has been measured for different energies of alpha particles, protons, and electrons.

## § 2. EXPERIMENTAL METHOD

The CsI(Tl) crystal was obtained from the Harshaw Chemical Company who stated that the molar concentration of thallium was 0.1%. The dimensions of the crystal were 1.5 cm  $\times$  0.9 mm and it was mounted by fine wires on a Du Mont 6292 photomultiplier parallel to the photocathode at a distance of 3.5 cm. A highly reflecting aluminium cylinder, 3.5 cm in diameter by 3.5 cm in length, with a very thin aluminium end window was used as a light reflector. This arrangement has been found to give good pulse height resolution for photomultipliers with photocathodes which are not quite uniformly sensitive.

The current pulses from the photomultiplier were integrated, the leakage time constant being 86  $\mu$ sec. The integrated current pulses, or voltage pulses, were then fed through a cathode follower and a matched delay line of 1.7  $\mu$ sec to a Tektronix 517A cathode-ray oscilloscope. Measurements were made on three time scales, 0.2  $\mu$ sec  $\text{cm}^{-1}$ , 0.5  $\mu$ sec  $\text{cm}^{-1}$  and 2  $\mu$ sec  $\text{cm}^{-1}$ . The pulses on the oscilloscope were photographed, projected on a large screen and carefully traced on fine graph paper.

The time base speeds on the Tektronix oscilloscope were compared with the period of a crystal controlled, standard frequency oscillator, Furzehill type G415. This was done by using the crystal controlled oscillator to check the frequency scale of a variable frequency oscillator, type Advance 62. Using convenient frequencies for each time base, the train of sine waves was randomly triggered, photographed and then projected and traced in the same way as the pulses from the CsI(Tl). The probable error in the calibration was  $\pm 1\%$ . The time base speeds indicated on the oscilloscope appeared to be systematically 2% higher than those given by our calibration. Our results have been corrected for this difference.

The electron source was the photopeak and Compton spectrum in the crystal for Cs<sup>137</sup> gamma rays ( $E = 661$  keV). A polonium source (5.3 MeV) was used at different distances from the crystal to give alpha particles of different energies. Higher energy alpha particles were obtained by bombarding boron 10 with 500 keV deuterons. From other experiments with a multichannel pulse height analyser, we have verified that most of the pulses in a limited region of the energy spectrum can be attributed to the reaction  $^{10}\text{B}(\text{d}, \alpha)^8\text{Be}$  ( $Q = 17.8, 14.91$  MeV). High energy protons were obtained from the reaction  $^{10}\text{B}(\text{d}, \text{p})^{11}\text{B}$  ( $Q = 9.23, 7.09, 4.77$  MeV).

Five different values of the photomultiplier voltage were used to make the pulse height on the oscilloscope about the same for 1 mev alpha particles, 4.8 mev alpha particles, 660 kev electrons, 2 mev protons and 8.5 mev protons. The change in photomultiplier gain at these different voltages was later measured using a Hutchinson-Scarrott 100-channel pulse height analyser. Knowing the differences in gain at the five voltages, all of the graphs of the voltage pulses could be normalized to the same gain.

### § 3. RESULTS

When the photographs of the voltage pulses were projected on a large screen, it was immediately obvious that the pulses due to alpha particles were rising more rapidly than those due to electrons, and that the rise time of the proton pulses lay somewhere between the two.

The graphs of the voltage pulses were analysed in two ways. In the first method, voltage pulse heights  $V_{1/2}$  and  $V_4$  were measured at  $0.5 \mu\text{sec}$  and  $4 \mu\text{sec}$ . These values were then normalized to allow for the changes in gain of the photomultiplier at the different voltages used in the experiments. When this was done, the values of  $V_{1/2}$  were plotted against  $V_4$ , and the result is shown in figure 1 in

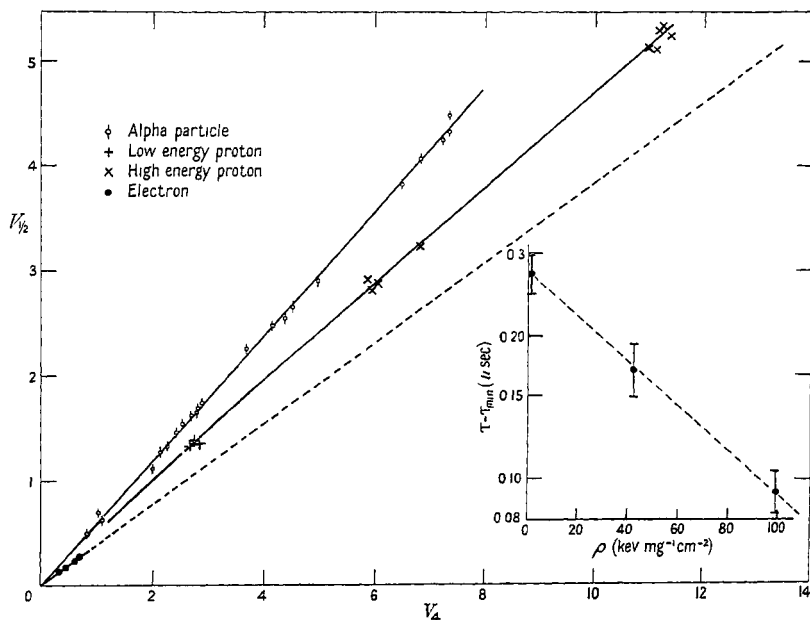


Figure 1. The minimum mass separation using CsI(Tl) as a single crystal detector.  $V_{1/2}$  and  $V_4$  are the voltage pulse heights at  $0.5 \mu\text{sec}$  and  $4 \mu\text{sec}$  respectively. Inset:  $\tau - \tau_{\text{min}}$  as a function of  $\rho$  the average ionization density.

which different symbols are used for points arising from different types of particles. It can be seen that there are three distinct loci for the points, one for each type of particle. The width of the loci can be calculated, if the number of photoelectrons released from the photocathode per unit proton energy is known. From the width of the peaks in the pulse height spectra measured on a multichannel pulse height analyser, we estimate this number to be greater than 5000 per 10 mev. Choosing

a fixed value of  $V_{1/2}$  for 10 mev protons, the percentage spread in the corresponding value of  $V_4$  is therefore about 1% since  $V_{1/2} \approx \frac{1}{2}V_4$ . Only pile up of pulses at very high counting rates can further increase the width of the loci. Figure 1 can be obtained directly using the X and Y plates of an oscilloscope and the mass separation can be further increased by a suitable choice of bandwidths of amplifiers for  $V_{1/2}$  and  $V_4$ .

In the second method of analysis, the graphs were corrected for the leakage time constant of  $86 \mu\text{sec}$ , and then carefully differentiated to obtain the shape of the current pulses. This was done for electrons of 660 kev, protons of 8.6 and 2.2 mev and alpha particles of 4.8 mev, two pulses being analysed in each case. The advantage of measuring the voltage pulse and then differentiating it, rather than measuring the current pulse directly is that it allows a flexible compromise between time resolution and charge fluctuation. When the derivatives of the voltage pulses were plotted, it was found that the current pulses rose with a time constant of  $50 \pm 10 \text{ m}\mu\text{sec}$ , which is attributed to the bandwidth of the delay cable and transit time variations in the Du Mont 6292. The effect of this rise time on the measured values of  $\tau$  was very small, but the initial decay rates were corrected to allow for its effect. All of the derived current pulses were then normalized to unity at time zero, and the results for the different particles plotted in figure 2. Values have not been plotted for the region  $1.6\text{--}4 \mu\text{sec}$  because residual mismatch of the delay line may render them uncertain.

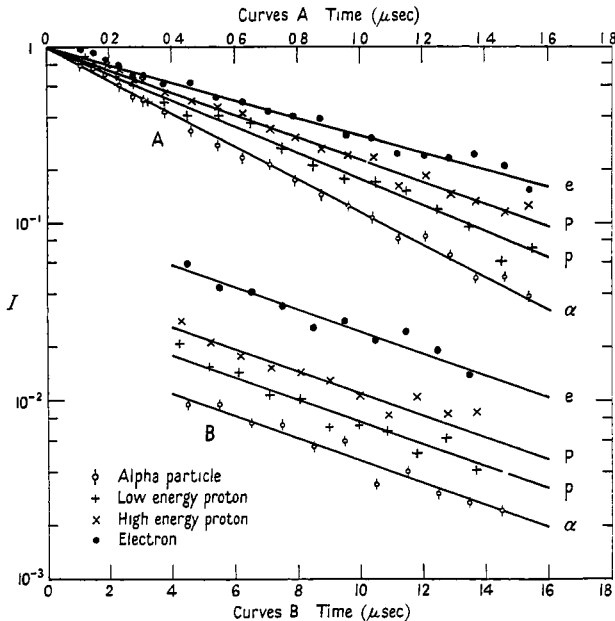


Figure 2. Decay curves in CsI(Tl) for different particles.

It can be seen from figure 2 that the decay of the current pulse can be conveniently described in terms of two components in each case, an initial fast decay and a slow component. If a third component exists with mean life of  $2\text{--}3 \mu\text{sec}$ , we estimate that it must contribute less than about 5% to the total voltage pulse at

17  $\mu\text{sec}$ . The subtraction of the common slow component which has a value of  $7 \pm 0.5 \mu\text{sec}$  from the initial decay shown in figure 2, gives the results shown in the table, for the average lifetime  $\tau$  during the time interval  $0.1 \mu\text{sec}$ – $1.6 \mu\text{sec}$ . For comparison, the energy of the particles used and the average ionization density  $\rho$  defined as the energy divided by the range, are also given in the table. The range for the electron taken from the empirical formula of Katz and Penfold (1952) has been increased by a factor of two to try to allow for electron scattering in the crystal.

The variation of  $\tau$  with  $\rho$  encourages us to believe that  $\tau$  may be a continuous function of  $\rho$ . Consideration of the number and accuracy of the points available does not permit us to derive the function very accurately. However the general behaviour is illustrated by the empirical formula  $\tau - \tau_{\text{min}} = (\tau_{\text{max}} - \tau_{\text{min}}) \exp(-\rho/\rho_0)$  which is suggested by the values in the table and also by the loci of figure 1. The inset in figure 1 is a logarithmic plot of  $\tau - \tau_{\text{min}}$  against  $\rho$ . The value of  $\rho_0$  obtained from this graph is  $\rho_0 = 95 \pm 15 \text{ keV mg}^{-1} \text{ cm}^{-2}$ , if  $\tau_{\text{min}} = 0.415 \mu\text{sec}$  and  $\tau_{\text{max}} = 0.685 \mu\text{sec}$ , our best estimates of  $\tau$  for alphas and electrons respectively. The values  $\tau_{\text{eqn}}$  in the table are calculated from the empirical formula using the above values.

Since all of our results could be normalized to the same electronic gain, we have been able to estimate the relative voltage pulse height per unit energy (or efficiency) for the different particles. Two efficiencies are given in the table:  $\epsilon_{1.5}$  is defined as the efficiency for light emitted in the time range up to  $1.5 \mu\text{sec}$ , and  $\epsilon_{\infty}$  is the efficiency for infinite time based on the assumption that there are no contributions from components longer than the common  $7 \mu\text{sec}$  component. Derived values of  $I_{\text{max}}/E$ , the relative peak currents per unit energy, and of  $R$  the ratio of the light output in the long component to the total light output are also given in the table.

	e	p	p	$\alpha$
Particle	Electron	Proton	Proton	Alpha
Energy $E$ (MeV)	0.66	8.6	2.2	4.8
$\rho$ (keV $\text{mg}^{-1} \text{ cm}^{-2}$ )	1.1	42.5	100	680
$\tau$ ( $\mu\text{sec}$ )	$0.70 \pm 0.025$	$0.60 \pm 0.02$	$0.52 \pm 0.01$	$0.425 \pm 0.01$
$\tau_{\text{eqn}}$ ( $\mu\text{sec}$ )	0.695	0.60	0.519	0.423
$\epsilon_{1.5}$	$1 \pm 0.05$	$1.56 \pm 0.08$	$1.49 \pm 0.08$	$0.75 \pm 0.04$
$\epsilon_{\infty}$	$1 \pm 0.05$	$1.18 \pm 0.05$	$1.04 \pm 0.05$	$0.48 \pm 0.03$
$I_{\text{max}}/E$	$1.0 \pm 0.1$	$1.75 \pm 0.2$	$1.9 \pm 0.2$	$1.15 \pm 0.1$
$R$	0.50	0.35	0.30	0.25

#### § 4. DISCUSSION

Our results show that in CsI(Tl) the efficiency and the decay time of the fluorescence vary with the density of ionization. It might be thought that both effects would be related and arise from the same cause. Kallmann and Brucker (1957) have shown that in a wide variety of scintillators which exhibit efficiency quenching effects which vary with ionization density there is no evidence for any variation in the decay time. Kallmann concludes that the main part of the quenching, at least in the organic scintillators, is finished before any appreciable amount of light is emitted. It might be expected that a similar fast quenching effect occurs to some extent in inorganic scintillators, but that in CsI(Tl) there may also be a slower mechanism for quenching, which competes with the process of emission during the time of emission. Since the efficiency of CsI(Tl) (and also LiI (Eu), Schenck and Neiler 1954) appears to first increase with ionization and then decrease, while

the fast quenching mechanisms observed in the organic crystals lead to a monotonic decrease in efficiency, it is necessary to look for a slow quenching mechanism, which will result in the efficiency increasing with increasing ionization density. If the observed variation  $\tau$  with ionization is attributed to a transport (or recombination) mechanism (Curran 1953), slow quenching occurring during the transport time to the emitting centres would result in a greater decrease in efficiency for smaller ionization densities, which correspond to longer decay times. Thus, qualitatively, this interpretation of our results would explain the observed variations of efficiency and lifetime with ionization density. Such a mechanism, including fast quenching, contains so many adjustable parameters that it is possible that the results for CsI(Tl) could be explained quantitatively, particularly as a transport mechanism would lead to a hyperbolic decay law which would be in rough agreement with our results. However, we have estimated transport or recombination times for CsI(Tl) and these are very much shorter than the observed decay times. Because of this fundamental discrepancy it seems unlikely that our results can be explained in terms of this mechanism.

Since the efficiency variation appears to depend mainly on the fast quenching which is expected to be independent of the emission process, we now go on to consider a possible interpretation of the variation of  $\tau$  with  $\rho$  independently of the variation of efficiency.

The dependence of  $\tau$  on so many different factors, and the limitations of the present experiments make it necessary to be cautious in comparing the observed variation of  $\tau$  with ionization density and the results of others in experiments on CsI(Tl) crystals. There is, however, a very striking similarity between the variation of  $\tau$  for alpha particles in CsI (1% Tl) as a function of temperature measured by Knoepfel *et al.* (1957) using an RCA 6655 and our own results for the variation of  $\tau$  with ionization density at constant temperature. Knoepfel *et al.* find that  $\tau$  for alpha particles decreases to a limiting value  $\tau_L$  as the temperature is increased to room temperature. We have plotted the logarithm of  $\tau - \tau_L$  as a function of temperature using their results, and find that

$$\tau - \tau_L \propto \exp(-T/T_0)$$

where  $T_0 = 13.5^\circ\text{C} \pm 1^\circ\text{C}$ . This empirical expression can be compared with our own results expressed as  $\tau - \tau_L \propto \exp(-\rho/\rho_0)$  where  $\rho_0 = 95 \pm 15 \text{ keV mg}^{-1} \text{ cm}^{-2}$ . The similarity of the two expressions, in each of which  $\tau_L$  is the value found for alpha particles suggests that the effect of changing the particle ionization density is to change the effective local temperature at which the emission takes place. If the change in local temperature is proportional to  $\rho$  then it is possible to deduce our results for different particles at the same temperature from those of Knoepfel *et al.* for the same particle at different temperatures.

Our results may be explained in terms of a simple model employing two sets of electron traps at different energy levels. Decay of the trap occurs when it receives enough thermal energy to reach an excited state, decay from which leads to fluorescence. It is supposed that the higher energy trap is responsible for the short lifetimes, while the lower energy trap gives rise to the  $7 \mu\text{sec}$  lifetime. The difference in the two lifetimes is due to the energy difference between the trap and the levels responsible for fluorescence. It would be expected that the trap with the smaller energy difference would be more susceptible to temperature changes than the other one. This would explain the difference in the short lifetimes for different particles if different values of  $\rho$  correspond to different

effective temperatures in the material surrounding the track, and would also account for the common long lifetime.

In the table  $R$  is the ratio of the light emitted due to the long lifetime ( $7 \mu\text{sec}$ ) to the total light emitted. It is seen that the shorter the initial lifetime, the less there is of the long component. This effect, also seen in figure 2, suggests that the lower energy trap is fed from the higher one in competition with the normal transition to the radiating state. In this case, the lifetime for decay from one trap to another must be of the order of  $1 \mu\text{sec}$ .

This interpretation of our results would lead us to expect a considerably greater change in the values of  $\tau$  for different particles in CsI(Tl) than are given here if the crystal temperature is lowered below room temperature. This result follows from the behaviour of  $\tau$  as a function of temperature observed for alpha particles by Knoepfel *et al.* and would result in a considerable increase in the particle separation shown in figure 1.

If this preliminary interpretation of our results is correct, it seems likely that similar effects may occur in other crystals in which non-radiating states are involved. It is possible, however, that it is only in CsI(Tl) that the characteristics of the resonance energy transfer are such as to result in a conveniently large variation in  $\tau$  as a function of available temperatures, and thus to a useful variation of  $\tau$  for different particles.

Another quite different interpretation of our results is that the variation in ionization density results in a different distribution in the initial population of radiating states. The observed variation in the value of  $\tau$  would then reflect the different contributions of many radiating states with different decay times. The simplest and most extreme case would be two radiating states corresponding to the values of  $\tau$  observed for alpha particles and electrons. It is not possible from our present results to deduce from the straightness of the log plots of the current pulses due to protons whether the values of  $\tau$  observed for protons are due to a combination of two lifetimes or one single lifetime. However, the order of magnitude of the values of  $\tau$  observed seems more consistent with those to be expected for traps rather than for radiating states.

A possible interpretation of the results given here could have been that the original decay of the main light pulse from the tracks of the particles is the same but complex. Since the path length of the light passing through the crystal to the photocathode increases in our experiments with increasing  $\rho$ , differential absorption of the longer components in the light pulse would tend to a shorter measured decay time for higher values of  $\rho$ . However, Wells (private communication) of this department, has found substantially the same values of  $\tau$  as we have for alpha particles and electrons in a different CsI(0.1%Tl) crystal, obtained from the Harshaw Chemical Company six months later than the one used in the experiments described here. The crystal which Wells used was about three times the thickness of our crystal and was mounted using silicone grease. In both cases we believe that the crystals were cut from the interior of a much larger block of material and therefore that the thallium concentration is uniform throughout our crystals.

It is hoped that further measurements, particularly on the emission spectrum for the different particles, will help to elucidate the mechanism involved. Further experiments will also be designed from the point of view of improving the mass separation.



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## REFERENCES

- BONANOMI, J., and ROSSEL, J., 1952, *Helv. Phys. Acta*, **25**, 725.  
CURRAN, S. C., 1953, *Luminescence and the Scintillation Counter* (London : Butterworths Scientific Publications).  
EBY, F. S., and JENTSCHKE, W. K., 1954, *Phys. Rev.*, **96**, 911.  
HUTCHINSON, G. W., and SCARROTT, G. G., 1951, *Phil. Mag.*, **42**, 792.  
KALLMANN, H., and BRUCKER, G. J., 1957, *Phys. Rev.*, **108**, 1122.  
KATZ, L., and PENFOLD, A. S., 1952, *Rev. Mod. Phys.*, **24**, 28.  
KNOEFFEL, H., LOEPFE, E., and STOLL, P., 1956, *Helv. Phys. Acta*, **29**, 241; 1957, *Z. Naturf.*, **12a**, 348.  
SCHENCK, J., and NEILER, J. H., 1954, *Nucl.*, **12** (3), 28.  
VAN SCIVER, W., and HOFSTADTER, R., 1951, *Phys. Rev.*, **84**, 1062.