Evidence for Positron-Electron Differences in Scintillation Response and Stopping Power*

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Positron-electron differences in scintillation response and stopping power of NaI(Tl) crystal have been studied using a two-crystal NaI(Tl) scintillation spectrometer. The scintillation response of the crystal to positron-electron pairs induced by gamma rays in the crystal was accurately measured. The energy distributions of positrons and of electrons induced in the crystal were calculated according to the formula of Nishina *et al.* In order to make clear the difference in scintillation responses to positrons and to electrons, gamma spectra were also measured by use of the same device. The scintillation response to positrons was obtained from the experimental response to positron-electron pairs and to electrons. The positron-electron difference in stopping power predicted by Rohrlich and Carlson has been confirmed with the present experiment. Although fairly good agreement with the theory can be seen at energies above 150 keV, the experimental positron-electron difference in stopping power in the lower energy range deviates from the theory to some extent.

1. INTRODUCTION

IN the past decade positron-electron differences in stopping power, energy straggling, multiple scattering, and range have been investigated by many workers. Rohrlich and Carlson[1] have calculated the stopping power and multiple scattering of positrons and electrons using the Bhabha and Møller cross sections. Numerical tables of stopping powers for positrons and for electrons have been calculated by Nelms[2] according to the formula of Rohrlich and Carlson. The calculated positron-electron difference of several percent in stopping power at energies below 100 keV may seem surprising. The difference would be, however, undetectable in experiments[3] concerned with the transmission of positrons and electrons through matter because of the quite large difference between positron and electron multiple scattering.[4] In the case of transmission measurements, positrons were found to be transmitted to greater extent than electrons except at low energies in aluminum.[5] In this case, electrons, being back-scattered to a greater degree than positrons, because of the cumulative effect in multiple scattering, were transmitted to a lesser degree. The results for aluminum, therefore, imply a larger stopping power for positrons than for electrons, but have not been conclusive as to the theoretical prediction on the positron-electron difference. Although this fundamental difference has been known for a decade, there seems to be little published literature concerning the experimental verification of this problem.

Recently, several experiments on the nonlinear response of NaI(Tl) crystals to various charged particles other than positrons have been reported.[6] The semi-empirical theory by Murray and Meyer[7] has shown that scintillation efficiency dL/dE of activated alkali iodides can be successfully predicted as a single smooth function of the specific energy loss of the incoming particles, dE/dX. The assumption 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 has been confirmed by Gwin and Murray.[8] The positron-electron difference in scintillation response of a NaI(Tl) crystal, however, has not yet been studied.

The purpose of this paper is (1) to show a positron-electron difference in the scintillation response of a NaI(Tl) crystal, and (2) to give some verification of the theoretical prediction of a positron-electron difference in stopping power. The scintillation response of the crystal to positron-electron pairs induced in the crystal was accurately studied using a two-crystal scintillation spectrometer. The energy distributions of positrons and of electrons induced in the crystal were calculated according to the formula of Nishina, Tomonaga, and Sakata.[9]

2. EXPERIMENTAL METHOD

A series of positron-electron pair-summing peak measurements was carried out using a two-crystal scintillation spectrometer, which is composed of a main spectrometer containing a 1-in.-diam×4-in.-long NaI(Tl) crystal (mounted on an RCA 6810A phototube), in conjunction with a side spectrometer containing a 3-in.-diam×2-in.-long NaI(Tl) crystal, to select

* Work partially supported by Japanese Atomic Energy Bureau.
[1] F. Rohrlich and B. C. Carlson, Phys. Rev. **93**, 38 (1954).
[2] A. T. Nelms, Natl. Bur. Std. (U.S.) Cir. No. 577 Suppl. (1958).
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[4] G. Groetzinger, W. Humphrey, Jr., and F. L. Ribe, Phys. Rev. **85**, 78 (1952); I. B. Strong and R. R. Roy, *ibid.* **131**, 198 (1963).
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[6] D. Engelkemeir, Rev. Sci. Instr. **27**, 589 (1956); P. Iredale, Nucl. Instr. Methods **11**, 336 (1961); W. Hink, Z. Physik **169**, 370 (1962).
[7] R. B. Murray and A. Meyer, Phys. Rev. **122**, 815 (1961).
[8] R. Gwin and R. B. Murray, Phys. Rev. **131**, 501 (1963).
[9] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, London, 1954), 3rd ed., p. 259; Y. Nishina, S. Tomonaga, and S. Sakata, Sci. Papers Inst. Phys. Chem. Res. (Tokyo) **24**, Suppl. No. 17 (1934).

the total absorption peaks of the annihilation radiation.

Gamma rays from Co^{60}, Na^{22}, Na^{24}, and ThC'' were collimated by 20-cm-long lead collimator as shown in Fig. 1. Signals from the main scintillation spectrometer were fed through a nonoverloading amplifier of the Fairstein type to an RCL 512-channel analyzer gated by the output of a fast-slow coincidence unit of about 100-nsec resolving time. Measurements were carried out several times for each positron-electron-pair energy. In order to avoid any nonlinearities which the amplifiers and the channel analyzer may have introduced, frequent calibrations were made with a specially stable pulse generator and with gamma rays from Na^{22}. During the experiment the room temperature was maintained at 24±1°C. Further, the over-all counting rate of the main scintillation spectrometer was kept below 2000 cps to avoid shifts throughout this experiment. To obtain large efficiency of coincidence counting, a two-crystal spectrometer was employed rather than the usual three-crystal spectrometer. One can not eliminate Compton-scattered peaks by use of the former arrangement, of course, but one can easily distinguish them from pair-summing peaks. Under these conditions

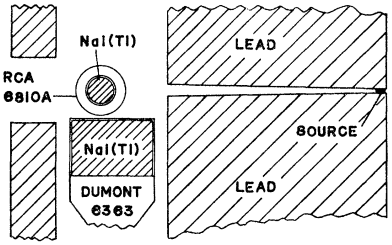

FIG. 1. Experimental arrangement. The collimating channel tapers from 1.0 cm diam at the crystal end to 0.3 cm diam at the source end.

the errors in estimation of pulse heights were likely to be less than ±1.5% in pair-summing-peak measurements, and ±0.75% in gamma-peak measurements. Figure 2 shows the positron-electron pair-summing spectra of Co^{60} gamma rays measured with the two-crystal spectrometer.

In order to make clear the difference in scintillation responses to positrons and to electrons, gamma spectra for monoenergetic gamma rays in the range 32 keV–2.75 MeV were also measured using the main scintillation spectrometer in the same geometry as described above.

The calibration by gamma rays can be expected to require correction from variations in light collection efficiency for energies below 100 keV. An experiment was carried out to establish the applicability of the work of Iredale on this effect.[10] A narrow, collimated beam of 0.66-MeV gamma rays from Cs^{137} was allowed to enter the main crystal so as to cross at various points having the same height, and the resulting pulse heights were carefully measured. The maximum differences in pulse height observed was 2%, as was the case in Iredale's work. His measured correction to the scintillation response was therefore applied: a 2% increase at 59.6 keV. Correction factors for gamma rays at energies above 59.6 keV were determined according to absorption coefficients of NaI for them.

[10] P. Iredale, Nucl. Instr. Methods **11**, 340 (1961).

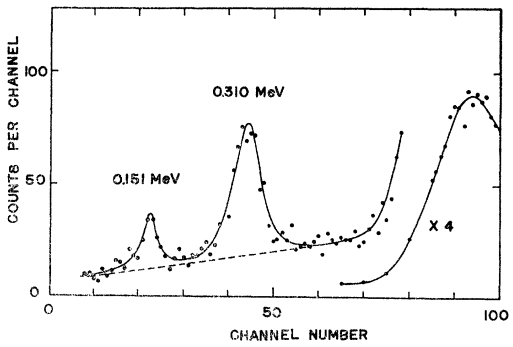

FIG. 2. Response of the two-crystal scintillation spectrometer to Co^{60} gamma rays. 0.151 MeV = (1.173–1.022) MeV single-pair summing peak, 0.310 MeV = (1.332–1.022) MeV single-pair summing peak, and big peak is Compton-scattered peak.

3. RESULTS AND DISCUSSION

Figure 3 shows the scintillation response of the NaI(Tl) crystal to positron-electron pairs and to gamma rays, in which the light output per MeV is plotted as a function of energy. The broken line indicates corrected values for light collection efficiency. In the low-energy range, the scintillation response to the former is considerably larger than to the latter having the same energy. However, there is little difference between them in the high-energy range. Results of measurements of gamma-ray spectra agree well with the results reported in Ref. 6. Therefore the experimental and calculated results of these other workers in the lower energy range were used for the calculations of L/E and dL/dE for electrons, although no attempt was made to extend the lower limit by the use of K x rays. The probability of annihilation before coming to rest in matter was neglected in this experiment because it is small.[11]

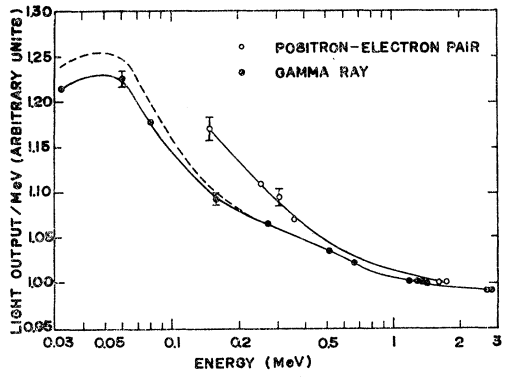

FIG. 3. Light output per MeV versus energy of positron-electron pair and gamma ray. The broken line indicates gamma-ray values corrected for light collection efficiency.

[11] J. W. M. Dumond, D. A. Lind, and B. B. Watson, Phys. Rev. **75**, 1226 (1949).

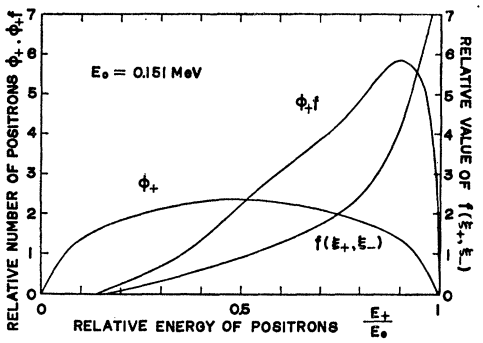

FIG. 4. The energy distribution ϕ_+f of positrons induced in NaI(Tl) by Co^{60}-1.173-MeV gamma ray. ϕ_+ indicates the energy distribution of positrons obtained by the application of the Born approximation. $f(\xi_+, \xi_-)$ indicates the correction factor introduced by Nishina *et al.*

In the calculation of the energy distribution of positrons formed in the crystal through pair creation, the energy distribution[12] ϕ_+ of positrons obtained by the application of the Born approximation was multiplied by the factor $f(\xi_+, \xi_-)$, introduced by Nishina *et al.*[9]:

$$f(\xi_+, \xi_-)=[2\pi\xi_+ 2\pi\xi_-]/[(e^{2\pi\xi_+}-1)(1-e^{-2\pi\xi_-})], \quad \xi_+=Ze^2/(\hbar v_+), \quad \xi_-=Ze^2/(\hbar v_-), \tag{1}$$

where Z is the nuclear charge, and v_+ and v_- are the velocities of positron and electron, respectively. In our case, $Z=53$ was used. A positron created in the Coulomb field of the nucleus is repelled and an electron is attracted by the nucleus. $f(\xi_+, \xi_-)$ causes a shift of the energy distribution in favor of larger positron energies. This shift of energy distribution is larger for lower positron-electron pair energies. The energy distribution of positrons is represented by $F(E_0, E_+)=\phi_+f(\xi_+, \xi_-)$, as shown in Fig. 4, where E_0 is the summation of energies of positron E_+ and electron E_-.The energy distribution of electrons is represented by $F(E_0, E_0-E_-)$. Figure 5 shows the calculated average energy of positrons created in NaI calculated; the average energy as a fraction of the energy of both particles is plotted as a function of the total energy.

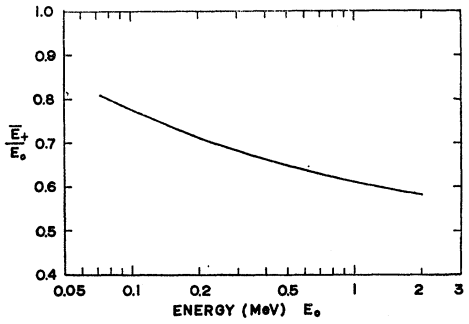

FIG. 5. The calculated average energies of positrons induced in NaI through pair creation. E_0 indicates the total energy of the positron-electron pair.

[12] W. Heitler, see Ref. 8, p. 258.

In order to obtain the scintillation response to positrons $L_+(E_+)$, the scintillation response to electrons $L_-(E_-)$ was obtained from the results of the measurements of gamma spectra according to the method of Zerby *et al.*[13]

The scintillation response to positron-electron pairs, $L(E_0)$, is the sum of the scintillation responses to positrons and electrons formed in the crystal. Therefore, one obtains the following equation:

$$L(E_0)=\frac{\int_0^{E_0} L_+(E_+)F(E_0, E_+)dE_+}{\int_0^{E_0} F(E_0, E_+)dE_+} + \frac{\int_0^{E_0} L_-(E_-)F(E_0, E_0-E_-)dE_-}{\int_0^{E_0} F(E_0, E_0-E_-)dE_-}, \tag{2}$$

where $L(E_0)$ is the scintillation response to a positron-electron pair whose energy is $E_0=E_++E_-$. As will be shown later, $L_+(E_+)/E_+$ and $L_-(E_-)/E_-$ are almost constant, within an error of $\pm 10\%$. Thus one may assume $L_+(E_+)=\alpha E_+$ and $L_-(E_-)=\beta E_-$ in the solution of the integral Eq. (2), and the expected error in the final result arising from this assumption will be less than 1%. Letting $L_+(E_+)=\alpha E_+$ and $L_-(E_-)=\beta E_-$, one obtains

$$L(E_0)=L_+(\bar{E}_+)+L_-(\bar{E}_-),$$

and

$$\frac{L(E_0)}{E_0}=\left(\frac{L_+(\bar{E}_+)}{\bar{E}_+}\right)\frac{\bar{E}_+}{E_0}+\left(\frac{L_-(\bar{E}_-)}{\bar{E}_-}\right)\frac{\bar{E}_-}{E_0}, \tag{3}$$

where $\bar{E}_+$ and $\bar{E}_-$ are the average energies of positrons and electrons, respectively. $L_+(E_+)/E_+$ was calculated from Eq. (3) using $L_-(E_-)/E_-$ as obtained above. Figure 6 shows the scintillation responses to positrons and to electrons; the light output per MeV is plotted as a function of energy.

dL/dE versus dE/dX for positrons and for electrons in NaI(Tl) crystal was obtained from the results shown in Fig. 6 according to the relation

$$dL/dE \equiv L/E+E(d/dE)(L/E). \tag{4}$$

The values of dE/dX for both particles were taken from the calculations of Nelms.[2] Figure 7 shows the scintillation efficiencies for positrons and for electrons; the broken lines show dL/dE for the corrected curve in Fig. 3, while the dotted line represents the semiempirical theory of Murray and Meyer.

The following causes may be considered for the dis-

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

agreement in dL_+/dE_+ and dL_-/dE_- for the same dE/dX in Fig. 7.

(a) Variation of light collection efficiency in the crystal. At energies below 100 keV a substantial part of the gamma rays interacting in the crystal are absorbed within the first millimeter of the crystal. Variation of light collection efficiency in the crystal, therefore, gives a smaller value for the scintillation response. The corrected values for the scintillation response and efficiency are shown by broken lines in Figs. 6 and 7.

(b) Multiple Compton scattering processes in the crystal. The disagreement in the high-energy range may be caused by multiple Compton scattering processes in the crystal. An increase of the response to gamma rays due to these processes can be estimated to be about 1% in our crystal from the scintillation efficiencies shown in Fig. 7.

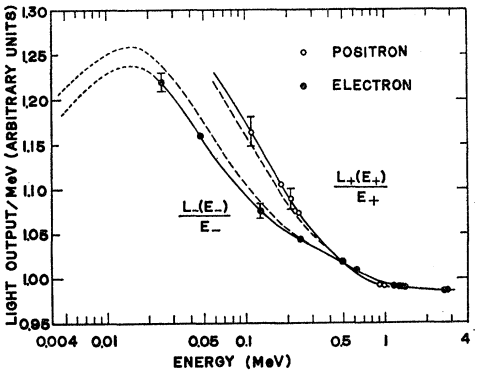

FIG. 6. Light output per MeV versus energy of positron and electron. The broken lines indicate light outputs for positrons and for electrons corrected for variation of light collection efficiency. The dotted portions of the curves at the low-energy end show the type of variation that was reported by Zerby *et al.*

(c) Inaccuracy in the energy distributions of positrons and of electrons. The inaccuracy in the energy distributions of positrons and of electrons may cause some change in the corresponding values of L/E. However, an error of 10% in the calculated average energy of the positrons causes only an error less than 1% in the scintillation response to positrons even in the low-energy range. Therefore this effect seems to be negligible.

(d) Inaccuracy in stopping powers. A larger stopping power for positrons than the stopping power calculated by Nelms could be expected in the low-energy range. According to the calculation by Nelms, dE_+/dX can be larger by 10% than dE_-/dX at low energy (40 keV) but smaller by about 3% in the high-energy range, 0.8–1.2 MeV. Assuming that the possible disagreement in the corrected curves results from a deviation of stopping power only, we have deduced the positron-electron difference in stopping power of a NaI(Tl) crystal. In Fig. 8 our experimental results for the positron-electron difference in stopping power of NaI are compared with the theoretical results.

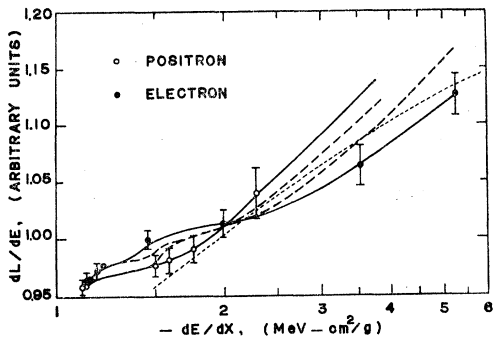

FIG. 7. Scintillation efficiency as a function of dE/dX as calculated by Nelms. The broken lines indicate scintillation efficiencies corrected for variation of light collection efficiency. The dotted line results from the semiempirical theory of Murray and Meyer. Vertical bars indicate the maximum errors.

A general feature of the energy dependence of the positron-electron difference in stopping power can be easily confirmed in Fig. 8. The error in dL_+/dE_+ is likely to be less than $\pm 2.5\%$ at the energy 110 keV. This error corresponds to an error of $\pm 20\%$ in stopping power. However, fairly good agreement with the theory can be seen at energies above 150 keV. The deviation from the theory at energies below 100 keV is ascribable to a possible incomplete correction for light collection efficiency.

4. CONCLUSION

The positron-electron difference in scintillation response has been observed in this experiment. At energies above 100 keV, agreement in dL/dE for two different particles (different in polarity of charge) having the same dE/dX has been shown within experimental errors. The energy dependence of the theoretical prediction for positron-electron difference in stopping power has been confirmed with the aid of the nonlinear response of the NaI(Tl) crystal to charged particles and the semiempirical theory of Murray and Meyer. Fairly good agreement with the theory can be seen at energies above 150 keV. The experimental positron-electron dif-

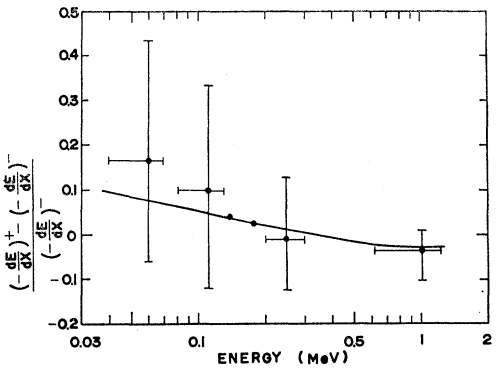

FIG. 8. Positron-electron difference in stopping power of NaI(Tl) as a function of the kinetic energy of particles in units of MeV. The solid line indicates the theoretical values. Vertical bars indicate the maximum errors.

ference in stopping power in the lower energy range, however, appears to deviate from the theory to some extent. Therefore it may be expected that the difference in stopping power is larger than the theoretical one calculated by Nelms below 100 keV. In order to clear up this point, a complete investigation of the stopping power of a NaI(Tl) crystal for positrons would be most desirable.

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Infrared Lattice Absorption by Gap Modes and Resonance Modes in KI

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Impurity-induced infrared absorption in KI containing Cl^- ions as substitutional impurities has been observed. A sharp line appears in the absorption spectrum in the neighborhood of 77 cm^{-1}, and at least three broad lines are observed near 61 cm^{-1}. The results of a theoretical calculation show that the identification of the former absorption peak with a localized mode whose frequency lies in the gap in the frequency spectrum of KI between the acoustic and optical branches is compatible with the experimental results. Similarly, the broad absorption peaks near 61 cm^{-1} can be identified with resonance modes in the acoustic continuum.

IT is our purpose in this note to present an experimental and theoretical demonstration of the existence of localized modes with frequencies in the gap between the acoustic and optical branches in the frequency spectrum of KI, as well as of resonance modes with frequencies in the acoustic continuum.

A powerful research tool for the study of the localized vibration modes introduced into the spectrum of crystal vibrations by impurity atoms has been infrared lattice absorption. Until very recently it has been used only in the investigation of the localized modes whose frequencies lie above the maximum frequency of the unperturbed host crystal.[1–7] It has been known for some time that if gaps occur in the frequency spectrum of a crystal it is possible for localized modes to exist whose frequencies lie in these gaps.[8] Moreover, it has been pointed out recently[9] that certain kinds of impurities, particularly very heavy ones, can give rise to resonance or quasilocalized modes whose frequencies lie in the range of normal-mode frequencies allowed to the perfect host crystal, and which are characterized by a greatly enhanced amplitude of vibration of the impurity atom.

The last two kinds of vibration modes should prove to be as interesting for theoretical and experimental study as the high-frequency localized modes. Up to the present time localized modes with frequencies in a

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