

FIG. 2. Microphotometer traces (redrawn) of  $\text{Sn}^{119m}$  and  $\text{Sm}^{153}$  spectra in the region from 19 to 42 keV. The similarity of the 19.8-keV line with other conversion lines (e.g., 21.2 and 36.1 keV) and the contrast with Auger lines are exhibited.

TABLE I. Intensities of conversion lines in  $\text{Sn}^{119m}$  electron spectrum.

Line	24.2- $L_I$	24.2- $M_I$	65.3- $K$	65.3- $L_{III}$	65.3- $M_V$
Energy (keV)	19.8	23.3	36.1	61.4	64.8
Density	$0.050 \pm 0.005$	$0.020 \pm 0.005$	$0.110 \pm 0.005$	$0.715 \pm 0.005$	$0.205 \pm 0.005$
Intensity (arb)	5.2	1.2	1.6	3.1	0.8

second. A crude estimate<sup>2</sup> of the slow neutron capture cross section of  $\text{Sn}^{118}$  for the production of  $\text{Sn}^{119m}$  was 0.004 barn. Much more intense sources have now been produced by irradiating a  $\text{Sn}^{118}$  sample (enriched to 91 percent) in a high neutron flux for a period of five months.<sup>3</sup>

Electron spectra of  $\text{Sn}^{119m}$  taken with 180° magnetic spectrographs are shown in Fig. 1. Two  $\gamma$ -transitions of  $24.2 \pm 0.5$  and  $65.3 \pm 0.5$  keV are observed. The energy of the former is computed on the basis of conversions mainly in the  $L_I$  and  $M_I$  shells. Also present in the spectrum is the well-known 390-keV  $\gamma$ -transition of  $\text{In}^{113m}$ , produced from  $\text{Sn}^{113}$ , which is itself produced from 0.2 percent of  $\text{Sn}^{112}$ . Although a lifetime check for a period of  $\sim 250$  days has not yet been made, the 24.2-keV transition is clearly identified as the second transition of  $\text{Sn}^{119m}$ , since it agrees well with the value of  $20 \pm 5$  keV determined recently for this transition by Scharff-Goldhaber *et al.*,<sup>4</sup> who used scintillation and proportional counters.

Since the conversion lines of the 24.2-keV transition lie within the region of the Auger lines from the Sn and In x-rays, it is important to verify that the 24.2- $L$  and 24.2- $M$  are not Auger lines. (From the intensity of the 390-keV conversion lines it can be shown that the In x-ray intensity, arising mainly from the 110-day  $K$ -capture activity of  $\text{Sn}^{113}$ , is approximately equal to the intensity of the Sn x-rays arising from conversion of the 65-keV  $\text{Sn}^{119m}$  transition. On this basis also, the capture cross section of  $\text{Sn}^{118}$  comes out to be 0.003 barn.) It should first be noted that the appearance of the 24.2- $L$  conversion line is different from that of an Auger group. This is clearly shown in Fig. 2, where the photometer traces of  $\text{Sn}^{119m}$  and  $\text{Sm}^{153}$  spectra, in similar energy regions, are compared. It is also clear from the intensity analysis that the 24.2- $L$  line is approximately 15 times more intense than that of all the indium Auger lines put together.

Measurement of the intensity of the 24.2-keV transition presents some difficulty on account of the low energies of the conversion lines. Previously, analysis of the intensity of such low energy lines has not been attempted because of the difficulty of correcting for photographic sensitivity and source absorption.

The values given in Table I have been derived using large corrections for photographic sensitivity, which are consistent with Cranberg and Halpern's values,<sup>5</sup> and also using Richardson's analysis<sup>6</sup> in order to correct for source thickness. These intensity values show that the 24.2- $L$  and 24.2- $M$  lines represent a transition which is approximately of the same intensity as the 65.3-keV transition. Theoretically, this should be the case, since the value of the  $L_I$  shell conversion coefficient for an  $M1$  transition of 24.2 keV is 7.5.<sup>7</sup> An indication that the corrections are of the right magnitude is shown by the  $L/M$  ratios for both transitions, which are  $\sim 4$ , a value which we have usually observed in previous work. The  $K/L$  ratio of the 65.3-keV transition is 0.51. This value is

lower than previously obtained and is in excellent agreement with Goldhaber and Sunyar's<sup>8</sup> empirical curves for an  $M4$  transition.

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<sup>1</sup> J. W. Mihelich and R. D. Hill, *Phys. Rev.* **79**, 781 (1950).

<sup>2</sup> J. W. Mihelich and R. D. Hill, *Phys. Rev.* **77**, 743 (1950).

<sup>3</sup> Enriched isotope obtained from Y12 plant, Oak Ridge, Tennessee, and irradiated by special arrangement with AEC, Isotopes Division, Oak Ridge, Tennessee.

<sup>4</sup> Scharff-Goldhaber, der Mateosian, Goldhaber, Johnson, and McKeown, *Phys. Rev.* **83**, 480 (1951).

<sup>5</sup> L. Cranberg and J. Halpern, *Rev. Sci. Instr.* **20**, 641 (1949).

<sup>6</sup> H. O. W. Richardson, *Proc. Phys. Soc. (London)* **63**, 234 (1950).

<sup>7</sup> Gellman, Griffith, and Stanley, *Phys. Rev.* **80**, 1028 (1950).

<sup>8</sup> M. Goldhaber and A. W. Sunyar, *Phys. Rev.*, to be published.

### Three-Quantum Decay of Positronium\*

MARTIN DEUTSCH

Department of Physics and Laboratory for Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts

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THE rate of decay by three-photon annihilation of ortho-positronium—the ground state of the bound electron-positron system with spin one—has been calculated theoretically with three different results: Ore and Powell<sup>1</sup> find  $\lambda_0 = 7.2 \times 10^6 \text{ sec}^{-1}$ , Lifshitz<sup>2</sup> gives  $\lambda_0 = 1.13 \times 10^7 \text{ sec}^{-1}$ , Ivanenko and Sokolov<sup>3</sup> obtain  $\lambda_0 = 1.6 \times 10^6 \text{ sec}^{-1}$ . Since the three papers start with the same physical assumptions, the discrepancy must be due to errors in at least two of the three calculations.

We have now determined this decay rate experimentally and find  $\lambda_0 = (6.8 \pm 0.7) \times 10^6 \text{ sec}^{-1}$ , in excellent agreement with the value calculated by Ore and Powell and in disagreement with the other authors. This seems to be the first case in which close experimental verification of a theoretical result has been possible for a third-order radiation process.

In a previous communication<sup>4</sup> we have reported the abundant formation of ortho-positronium in some gases and the action of nitric oxide in low concentration to destroy it by rapid conversion to the para state. This property of NO permits us to separate effects due to ortho-positronium from other phenomena. By this means we have found that in dichlorodifluoromethane ("Freon 12") at pressures above about 0.4 atmosphere substantially all annihilation processes occurring with delays exceeding  $8 \times 10^{-8} \text{ sec}$  are due to ortho-positronium. At lower pressures a second group appears which is not suppressed by NO and shows a rapid decay depending strongly on pressure. We make the hypothesis that this behavior is due to a large "positron attachment coefficient" of  $\text{CCl}_2\text{F}_2$  which results in an anomalously large molecular annihilation cross section for free positrons. It may not be accidental that Freon is known to show extremely strong electron attachment.

Whatever the exact mechanism for suppressing the free posi-

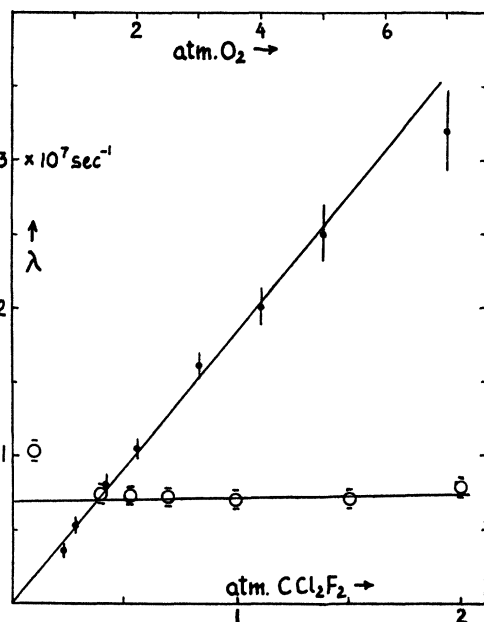


FIG. 1. Rate of decay of annihilation radiation in  $\text{CCl}_2\text{F}_2$  (open circles) and in  $\text{O}_2$  (full circles). The pressure scales are adjusted so that the abscissa corresponds to the same electron density in both gases.

trons, the delayed annihilation radiation in Freon at moderately high pressure seems to be due entirely to ortho-positronium. Its decay rate should depend somewhat on gas pressure  $p$  so that  $\lambda = \lambda_0 + ap$ . The pressure proportional term represents annihilation in collisions with Freon or impurity molecules.

The method used to measure the decay rate was the same as in the previous communication.<sup>4</sup> Figure 1 shows the results as a function of gas pressure. The straight line represents the equation  $\lambda = 6.8 \times 10^6 + 0.3 \times 10^6 p$ . The value  $\lambda_0 = 6.8 \times 10^6 \text{ sec}^{-1}$  for the rate of three-quantum decay, obtained by extrapolating the line to zero pressure, should be good to ten percent. The coefficient  $a = 0.3 \times 10^6 \text{ sec}^{-1} \text{ atm}^{-1}$  is rather uncertain. In any case,  $a$  is small, in agreement with calculations of Ore concerning the great stability of positronium against collisions in most gases. The point in Fig. 1 at 0.1 atmosphere which does not fall on the straight line is included to show the effect of free positrons at low pressures.

For comparison, Fig. 1 also shows the decay rate of annihilation radiation in oxygen as a function of pressure. In this gas, at pressures above about 0.5 atmosphere all positronium is converted to the para state so rapidly that substantially all delayed annihilation radiation is due to free positrons decaying in collisions. The rate of decay is, therefore, directly proportional to gas density as shown in Fig. 1. A more detailed discussion of both curves will be given in a later communication.

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<sup>1</sup> A. Ore and J. L. Powell, *Phys. Rev.* **75**, 1696 (1949).

<sup>2</sup> E. M. Lifshitz, *Doklady Akad. Nauk S.S.S.R.* **60**, 211 (1948).

<sup>3</sup> D. Ivanenko and A. Sokolov, *Doklady Akad. Nauk S.S.S.R.* **61**, 51 (1948).

<sup>4</sup> M. Deutsch, *Phys. Rev.* **82**, 455 (1951).

## A Tentative Interpretation of the Second Maximum in the Transition Curve for Cosmic-Ray Showers

TSAI-CHÜ

Observatoire du Pic du Midi, Bagnères-de-Bigorre, France

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THE experimental results on the second maximum still remain in controversy. Bothe *et al.*<sup>1,2</sup> have repeatedly reported the second maximum in the transition curve at 15-cm Pb. While

others found a small hump not necessarily at the same place or even nothing at all, formerly Clay,<sup>3</sup> and recently Kameda and Miura,<sup>4</sup> Fenyves and Haiman,<sup>5</sup> and Chaudhary<sup>6</sup> confirmed its existence. The explanation probably lies in the interpretation of this phenomenon, and the discrepancy in experimental results arises possibly from the differences in the arrangement of counters<sup>6</sup> or in the sensitivity of counters to low energy photons.

The nature of primary and secondary particles of the narrow showers responsible for the second maximum remains unknown. Schmeiser and Bothe<sup>7</sup> found that the second maximum is more prominent in the basement than under the roof. Absorption measurements<sup>7</sup> on the secondary particles produced in 1.5 cm Pb as well as in 15 cm Pb show that their coefficients of absorption are much smaller than those of electrons; the particles from 1.5 cm Pb are even more penetrating. In adjacent absorbers these penetrating particles produce further soft showers, the so-called *Zusatzstrahlung*.<sup>8</sup> Bothe<sup>1</sup> considered the narrow showers as the production of mesons by mesons, a new process not yet definitely confirmed by other experiments. In a sub-basement under about 60 cm concrete and with 15 cm Pb above a cloud chamber, Shutt<sup>9</sup> observed many showers containing two penetrating particles making an angle around 6 degrees, the frequency being 1 shower for every 4000 single penetrating particles; Jánossy<sup>10</sup> gave 1 in 12,000. The frequency of a meson accompanying a knock-on electron, which constitutes essentially the background of the transition curve, is 6.9 per 100 mesons<sup>11</sup> near sea level. Therefore, in a cloud chamber the knock-on phenomenon is at least 280 times more frequent than a penetrating pair originating from the lead above it. The selection by counters due to absorption and arrangements reduces the knock-on electrons, but a maximum as large as 35 percent (curve C<sup>2</sup>)—that means the frequency of penetrating showers is increased about one hundred times by counters—can never be expected due to penetrating showers. Furthermore, a small maximum probably exists in the transition curve (curve d<sup>2</sup>) for non-ionizing primaries; about 65 percent of narrow showers from 1.5-cm Pb are produced by non-ionizing primaries<sup>8</sup> and 25 percent from 15-cm Pb;<sup>8</sup> these facts quickly rule out the meson as the primary.

Kameda and Miura<sup>4</sup> proposed nucleons as primaries. Small angle showers are less penetrating, according to theory and experiment, but frequency considerations still render this proposal impossible. The nucleon intensity decreases much faster than the knock-on electrons in the underground, while the secondary mesons are not shower-producing. Transition curves for penetrating showers are either saturated at great thicknesses<sup>10</sup> or even increase appreciably;<sup>9</sup> no experiment<sup>12</sup> gives a sharp maximum. Only the soft components associated with the penetrating particles, such as knock-on electrons and gamma-radiations in nuclear phenomena, could probably produce a maximum. The striking difference<sup>8</sup> in the contribution of ionizing and non-ionizing primaries at 1.5 and 15 cm Pb cannot be understood with the known properties of protons and neutrons. Because nucleons have a comparatively long mean free path for showers of small multiplicity,<sup>13</sup> it is surely impossible that there are more penetrating showers at 1.5 cm Pb than at 15 cm.

It seems that narrow showers are intimately connected with the soft component. The only thing necessary is to explain the abnormal coefficient of absorption and the possibility of a second maximum appearing. Without introducing new unstable particles,<sup>6</sup> I have tentatively applied the idea of Cocconi and Greisen<sup>14</sup> to explain a small maximum<sup>15</sup> (it may be called rather an irregularity at present) between 5 and 10 cm in the transition curve. A similar irregularity in the same region was present in the curve of Altmann, Walker, and Hess,<sup>16</sup> and also in the Hg curves of Clay.<sup>3</sup> Cascade showers die out by leaving a large number of photons of the order of critical energy, their minimum coefficient of absorption, 0.19 per radiation length (or 1/2.7 per cm lead), being much smaller than that of electrons. The surviving photons degrade their energy mainly by the Compton process, and less frequently by pair production. Some electrons produce further brems-