

# Monoenergetic Positron Line and Three New Gamma Transitions in Bi<sup>206</sup>†

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The monoenergetic positron line (the 1.72-MeV transition) in Bi<sup>206</sup> was sought using an intense Bi<sup>206</sup> source in a two-cycle baffled solenoid spectrometer. The source was prepared by bombarding in a deuteron beam a highly purified radiogenic lead target with Pb<sup>206</sup> isotopic content enriched from 26 to 87.7%. The comparative Bi<sup>205</sup> contamination is thus reduced to less than 0.05%. The leak through of the opposite sign of particles of the baffle used is better than 1 in 10<sup>7</sup>. A small peak of ~1% above the base line is observed at the predicted momentum value for the monoenergetic positron line (3970 Gcm). The intensity of the positron line is estimated to be 3.2×10<sup>-3</sup> per gamma ray, which is one-sixth of that first reported by Brunner. The retardation factor of this *E1* (1.720 MeV) transition is estimated to be around 600. Three new gamma transitions above the highest energy line (1.720 MeV) known previously have been found and identified. These are 1.845, 1.88, and 1.90 MeV. From the ratios of internal conversion to internal pair production, one can assign *E1* to both 1.845- and 1.880-MeV transitions. The 1.90-MeV transition is probably also of the *E1* type.

## INTRODUCTION

IN 1953, Sliv<sup>1</sup> predicted that there, in fact, should exist an effect in which monoenergetic positrons are emitted. This process would occur when a nucleus is excited by more than  $2mc^2$  and when, at the same time, there is a vacancy in one of the inner electronic shells of the atom, to which the nucleus belongs. The nucleus can then de-excite in the following way: An electron-positron pair is created and the electron gets trapped in the hole of the atomic shell but the positron is emitted. If  $E_{em^+}$  is the kinetic energy of the positron then

$$E_{em^+} = E_\gamma - 2mc^2 + E_B,$$

where  $E_\gamma$  is the excitation energy of the nucleus and  $E_B$  the binding energy of the electron in the atomic shell. The ratio of the probability for the emission of monoenergetic positrons to that for the emission of  $\gamma$  rays is given by the following formula:

$$\frac{N_{em^+}}{N_\gamma} = \alpha_{em^+} P_v \left( \frac{\tau_v}{\tau_v + \tau_n} \right). \quad (1)$$

Here  $\alpha_{em^+}$  is the probability for emission of a monoenergetic positron per emitted  $\gamma$  ray in case that the nucleus belongs to an atom with a permanent vacancy in one of the electronic shells.  $P_v$  is the probability that the excited nucleus is surrounded by an electronic vacancy,  $\tau_v$  is the lifetime of the vacancy, and  $\tau_n$  is the nuclear lifetime.  $P_v$  depends on how the nuclear level is excited. It will be different from zero in several cases such as electron capture,  $\alpha$  decay, nuclear reactions, and preceding conversion electron transitions. It will, however, be of the order of magnitude of 1 only in the cases of electron capture and of preceding highly converted electromagnetic transitions. Here a vacancy

in the *K* shell will be the most probable one unless the transition energy is smaller than the *K* binding energy. For this reason only *K*-shell vacancies will be considered from here on.

While  $\tau_v^K$  is between 10<sup>-15</sup> and 10<sup>-17</sup> sec for medium- and high-*Z* atoms (the *Z* dependence is *Z*<sup>-4</sup> approximately)  $\tau_n$ , as given by the single-particle model, approaches the above time region only in the cases of *E1* and *M1* transitions. Even for these transitions usually  $\tau_v^K \lesssim \tau_n$  and consequently  $N_{em^+}/N_\gamma$  will depend on the ratio  $\tau_v^K/\tau_n$ . Since  $\tau_v^K$  is well known from measurements of the width of x-ray lines, a determination of  $N_{em^+}/N_\gamma$  will give an estimate for  $\tau_n$ , assuming that  $\alpha_{em^+K}$  and  $P_v^K$  in (1) are known. It might be worthwhile to point out that except for resonance fluorescence, which is limited to ground-state transitions, no other experimental methods are available for the measurements of nuclear lifetimes in the region 10<sup>-15</sup>-10<sup>-17</sup> sec.

$\alpha_{em^+K}$  was calculated first by Sliv<sup>1</sup> and later by Lombard and Rys.<sup>2</sup> Sliv evaluated  $\alpha_{em^+K}$  as a function of energy for *E1* and *E2* transitions. He used *Z*=83 and *Z*=55 for *E1* and *Z*=83 for *E2* (Fig. 1). Sliv also obtained the dependence of  $\alpha_{em^+K}$  on *Z* for *E1* and *M1* transitions (Fig. 2). Lombard and Rys calculated  $\alpha_{em^+K}$  as a function of energy in the case of *E1* transitions. This was done for three different values of *Z*, namely, *Z*=30, *Z*=63, and *Z*=82 (Fig. 3). A comparison of the curve obtained for *Z*=82 with the one given by Sliv for the multipolarity of *E1* and *Z*=83 shows a discrepancy which is attributed to an inadequate mathematical approximation in Sliv's work.

As can be seen from Figs. 1 and 2,  $\alpha_{em^+K}$  is largest for *E1* transitions. The same is true for  $\tau_v^K/(\tau_v^K + \tau_n)$ . It, therefore, becomes clear that these transitions are the most favorable ones in search for monoenergetic positrons. Another requirement is that the isotope must be in the medium- or high-*Z* region. This is necessary because of the limitation on energy resolution. The

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<sup>1</sup> L. A. Sliv, Zh. Eksperim. i Teor. Fiz. **25**, 7 (1953); J. Phys. Radium **16**, 589 (1955).

<sup>2</sup> R. Lombard and F. Rys, Nucl. Phys. **31**, 163 (1962).

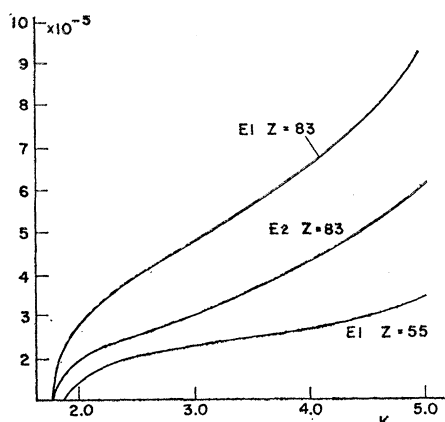


FIG. 1.  $\alpha_{e_m}^{+K}$  as a function of energy for  $E1$  and  $E2$  transitions (reference 1).

energy of the positron line has to be sufficiently different from the maximum energy of the continuous distribution of positrons from internal pairs, since the pairs are always competing with monoenergetic positrons. In other words,  $E_B^K/(E_\gamma - 2mc^2)$  has to be a few times larger than the instrumental energy resolution. Another experimental limitation may lie in the presence of other positrons either from  $\beta^+$  decay accompanying  $K$  capture or from pairs of higher energy electromagnetic transitions.

In 1959, Brunner, Leisi, Perdrisat, and Scherrer<sup>3</sup> published their results on the presence of a positron line in  $\text{Bi}^{206}$ . This isotope decays by  $K$  capture to an excited 5- level of 3.403 MeV in  $\text{Pb}^{206}$ , which in turn de-excites by several  $\gamma$  rays to the ground state. The most energetic one is a 1.72-MeV  $E1$  transition leading to the 1.684-MeV 4+ state in  $\text{Pb}^{206}$ .<sup>4</sup> Brunner *et al.* found that

$$(N_{e_m}^{+K}/N_\gamma)_{1.72 \text{ MeV}} = 2.6 \times 10^{-7}.$$

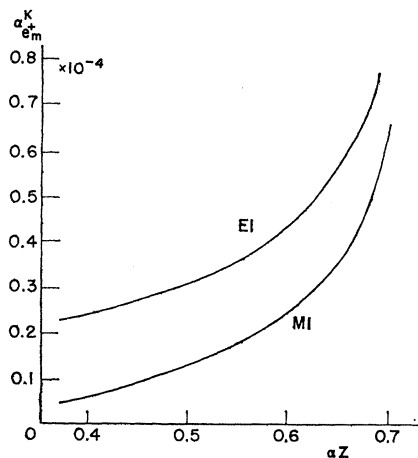


FIG. 2.  $\alpha_{e_m}^{+K}$  as function of  $Z$  for  $E1$  and  $M1$  transitions (reference 1).

<sup>3</sup> T. H. Brunner, H. J. Leisi, C. F. Perdrisat, and P. Scherrer, *Phys. Rev. Letters*, **2**, 207 (1959); H. J. Leisi, J. H. Brunner, C. F. Perdrisat, and P. Scherrer, *Helv. Phys. Acta*, **34**, 161 (1961).

<sup>4</sup> D. E. Alburger and M. H. L. Pryce, *Phys. Rev.* **95**, 1482 (1954).

Using the  $(\alpha_{e_m}^{+K})_{1.72 \text{ MeV}}$  given by Sliv for  $Z=83$  and the multipolarity  $E1$  and taking  $P_\nu^K=0.77$  they concluded that the 1.72-MeV  $E1$  transition in  $\text{Pb}^{206}$  is retarded only by a factor of 50 in comparison with that calculated from the single-particle model. Since this factor is much smaller than any other known for  $E1$  transitions in the medium- and high- $Z$  region<sup>5</sup> we decided to reinvestigate the presence and the intensity of the monoenergetic positron line found in the decay of  $\text{Bi}^{206}$ .

## EXPERIMENTAL METHOD

The experimental method used for the detection of monoenergetic positrons was straightforward: An accurate measurement of the positron spectrum of  $\text{Bi}^{206}$  using a magnetic spectrometer. In view of the very low intensity expected for the positron line there are, however, several requirements which are essential for the successful performance of the experiment:

1. The source has to be extremely strong (10–100 mC), yet thin and free of any radioactive contaminations.

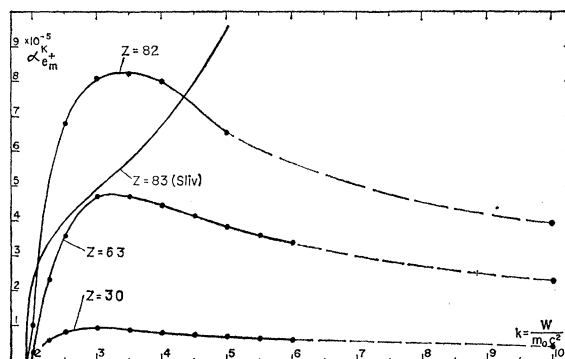


FIG. 3.  $\alpha_{e_m}^{+K}$  as function of energy for  $E1$  transitions (reference 2).

2. The spectrometer has to have good electron-positron discriminating baffles; i.e., the electron-positron rejection ratio must be extremely high.

3. Background has to be low. Here background includes two different kinds; background coming from cosmic rays and radioactivity of the surroundings (source-independent background) and background from the source, i.e.,  $\gamma$  rays and secondary electrons scattered into the detector by the walls of the spectrometer (source-dependent background).

4. Stable detector.

The investigation was carried out in two independent runs. In the first run, the source used was prepared by deuteron bombardment on spectroscopically pure natural lead. Later on in the second run, the experiment was greatly improved with a source prepared from radio-

<sup>5</sup> M. Deutsch and O. Kofoed-Hansen, in *Experimental Nuclear Physics*, edited by E. Segrè (McGraw-Hill Book Company, Inc., New York, 1959), Vol. III, p. 402.

genic lead where the isotopic concentration of  $\text{Pb}^{206}$  is as high as 88%. The strength of the source increased by a factor of 6 and the amount of the major impurity,  $\text{Bi}^{205}$ , was reduced 5 times. The results obtained from the second experiment are therefore cleaner and more accurate. However, in order to describe the different methods which were used for the preparation of the source and to show the consistency in the results obtained, both runs of the experiment will be described in the following paragraphs.

#### SOURCE PREPARATION

$\text{Bi}^{206}$  is obtained from the  $\text{Pb}^{206}(d,2n)\text{Bi}^{206}$  reaction. By bombarding a lead target with deuterons the following additional reactions take place, leading to positron emitters, which would interfere with the measurement of the positron line in  $\text{Bi}^{206}$ :

$$\text{Pb}^{204}(d,2n)\text{Bi}^{204}_{12h}, \quad Q \approx -7.5 \text{ MeV},$$

$$\text{Pb}^{204}(d, n)\text{Bi}^{205}_{14d}, \quad Q = 1 \text{ MeV},$$

$$\text{Pb}^{206}(d,3n)\text{Bi}^{205}_{14d}, \quad Q = -17 \text{ MeV}.$$

Since the  $Q$  value of  $\text{Pb}^{206}(d,2n)\text{Bi}^{206}$  is  $-8$  MeV, the last of the above reactions can be avoided by lowering the deuteron energy to less than 17 MeV. The isotopic composition of natural and radiogenic lead is given in Table I. From this table, one can see that the two

TABLE I. Isotopic composition of natural and radiogenic lead.

Isotope	Isotopic conc. in natural lead (%)	Isotopic conc. in radiogenic lead (%)
$\text{Pb}^{204}$	1.3	<0.1
$\text{Pb}^{206}$	26	88
$\text{Pb}^{207}$	21	8.9
$\text{Pb}^{208}$	52	3.4

reactions starting from  $\text{Pb}^{204}$  can be minimized by the use of radiogenic lead.

The deuteron bombardments were made in the Brookhaven cyclotron, where the energy of the deuterons is about 21 MeV. By use of an aluminum degrading foil this energy was reduced to 15 MeV. Then, with a target thickness of 0.01 in. the yield of the  $\text{Pb}^{206}(d,2n)\text{Bi}^{206}$  reaction was  $65 \mu\text{C}/\mu\text{A h}$  for natural lead and  $200 \mu\text{C}/\mu\text{A h}$  for radiogenic lead. The bombarding time was 20 h and the average current  $50 \mu\text{A}$  in both cases.

Two different methods for separating bismuth from lead were used.

1. A "filter method" based on the difference in the solubilities of bismuth and lead compounds, such as  $\text{Bi}(\text{OH})_3$ ,  $\text{Pb}(\text{OH})_2$ ,  $\text{BiCl}_3$ , and  $\text{PbCl}_2$ . This method is described elsewhere.<sup>6</sup> After having repeated the pro-

cedure twice the radioactive bismuth was electroplated out of a HCl solution onto a  $\frac{1}{8}$ -in.-diam circle of a copper foil with a thickness of 0.001 in. The amount of solid materials plated out of the solution together with the bismuth was less than  $100 \mu\text{g}$ , and the source thickness was smaller than  $0.7 \text{ mg}/\text{cm}^2$ . The over-all efficiency for the source preparation was 15–20%.

2. An ion-exchange method, described by Nelson and Kraus.<sup>7</sup> Here the target was converted into the nitrate and the solution passed through an ion-exchange column in a 0.05N HCl solution. After being eluted from the column in 1M  $\text{H}_2\text{SO}_4$  the bismuth was again

TABLE II. Source specifications.

	Source 1	Source 2
Target comp.	natural lead	radiogenic lead
Bi-Pb sep.	filter method	ion-exchange method
Strength	13 mC	80 mC
$\text{Bi}^{205}$ content	0.25%	0.05%

plated onto a  $\frac{1}{8}$ -in. diam of a 0.001-in. copper foil. This method turned out more successful than the "filter method", from the point of view of over-all efficiency (30–35%), final source thickness, and preparation time.

Table II shows the specifications of the two  $\text{Bi}^{206}$  sources which were prepared for the experiment.

The percentage of  $\text{Bi}^{205}$  present in the source was measured by the intensity of the 1.766-MeV  $K$ -conversion line of this isotope. Figure 5 shows the electron spectrum in the vicinity of this line as it was obtained from source 2 at a time when the latter was 3 weeks old.

#### SPECTROMETER

The iron-free, homogeneous field spectrometer of Columbia University shown in Fig. 6 was used to measure the positron and electron spectra of  $\text{Bi}^{206}$ . This spectrometer has several features which make it particularly suitable for this investigation.

1. Since it is iron-free there is no difference in energy calibration between electron and positron lines.

2. It has a two-cycle baffle, which minimizes source-dependent background.

3. It has excellent electron-positron discriminating baffles. Instead of the spiral baffle frequently used in lens-type spectrometers a different kind of baffle was designed especially for this experiment. It contained much less mass than the spiral one and therefore caused less scattering of electrons and  $\gamma$  rays. It was made in three sections as shown in Figs. 6(a) and 6(b). Two 75-deg segments were placed opposite each other in a plane perpendicular to the axis of the spectrometer near the source. Two similar segments were placed in the same way one third of a cycle further down on the axis, displaced by rotation of 60 deg around the axis. A third set of segments, again displaced by 60 deg,

<sup>6</sup> Atomic Energy Research Establishment Lib/Trans 722 (unpublished), p. 56.

<sup>7</sup> F. Nelson and K. A. Kraus, J. Am. Chem. Soc. **76**, 5916 (1954).

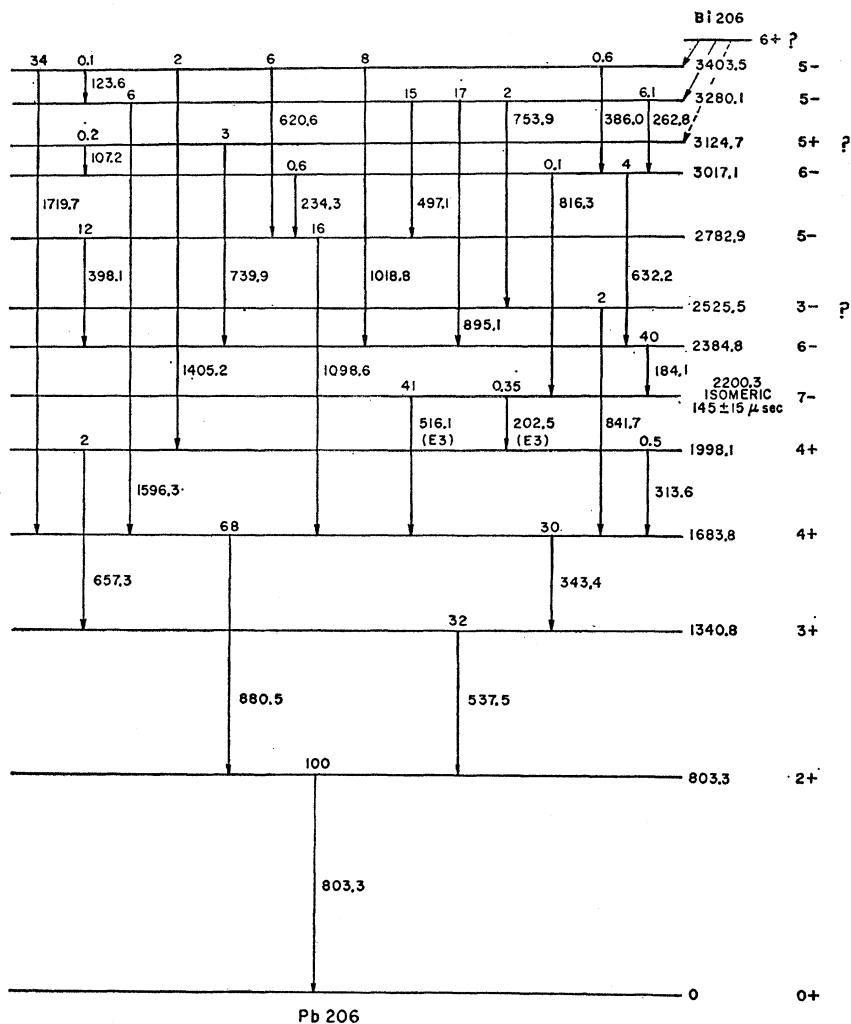


FIG. 4. Decay scheme of  $\text{Bi}^{206}$ , taken from Alburger and Pryce (reference). The number at the head of each transition is its intensity in percent per disintegration.

was placed after another third of a cycle further down on the axis. Note that all three sets of segments are within the first cycle. Particles of the right sign charge spiralling down along the axis of the spectrometer will be transmitted by the second and third section baffles, while particles of the wrong sign charge will be stopped by them. Theoretically each part would only need to occupy an angle of 60 deg but the overlap of 15 deg makes the adjustment of the baffles less critical and improves the electron-to-positron-rejection ratio. The transmission of the baffles is reduced approximately to  $210/360 = 58\%$  of its original value.

The electron-to-positron-rejection ratio was measured by use of a  $\text{Cs}^{137}$  source with a strength of 10 mC. First a  $40\text{-}\mu\text{C}$   $\text{Cs}^{137}$  source was put into the spectrometer and the  $K$ -conversion line of its 0.667-MeV transition was measured. Then the  $40\text{-}\mu\text{C}$  source was replaced by the 10-mC one and the current through the spectrometer coils switched into the opposite direction. Careful measurements were made especially for values of current for which the 0.667-MeV  $K$ -conversion elec-

tron line was observed. Figures 7 and 8 show the result. There is no indication of any peak and the value  $10^{-7}$  is obtained from the statistical uncertainty for the electron to positron rejection ratio, assuming that the electrons originate from a line. There seems, however, to be some leak through of electrons below the value of current, which corresponds to the end point of the continuous  $\beta^-$  spectrum of  $\text{Cs}^{137}$ . This is also demonstrated by the fact that a different spectrum was obtained when the source was covered by a  $\frac{1}{4}$ -in.-thick Lucite absorber. This leak through of inelastically scattered electrons does not interfere with the measurement of a positron line, since it will not show up as a line. It will, however, make the detection of very weak continuous  $\beta^+$ -spectra more difficult, if these spectra come from sources emitting electrons in the same or higher energy region.

#### DETECTOR

As was already pointed out before, a low-background stable detector is essential for the measurement of a

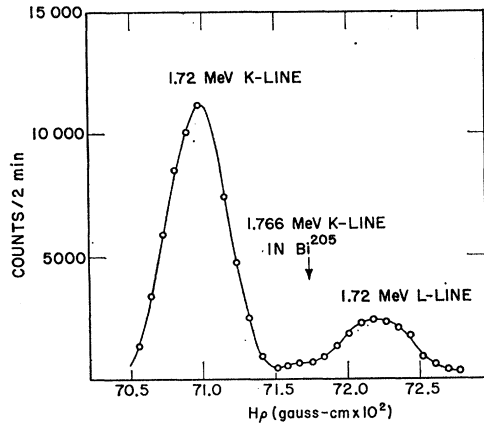


FIG. 5. Electron spectrum around 1.7 MeV of 80-mC  $\text{Bi}^{206}$  source, 3 weeks old.

positron line. For the sake of stability a Geiger counter is desirable. Background will be lowest if its volume is kept as small as possible, i.e., just large enough to receive all the focused particles. In the spectrometer used an electron beam from a point source gets focused to a line, which after two cycles has the length of 2 in. In case of an extended source the image at the focus will have the area of the source and will be 2 in. long. The beam hits the axis of the spectrometer at an angle of about 40 deg. Then small-volume Geiger counters of two kinds can be used. One possibility is a side-window counter of cylindrical shape, whose diameter is equal to or slightly larger than the diameter of the source, and whose length is slightly smaller than 2 inches. It has to be placed along the spectrometer axis at the line focus. The other possibility is a flat end-window counter (so called pancake counter), placed with its window perpendicular to the spectrometer axis at the middle of the line focus. For maximum efficiency its window has to have a diameter slightly smaller than 2 in. A counter of the first kind was made especially for the purpose of the experiment. It is shown in Fig. 9.

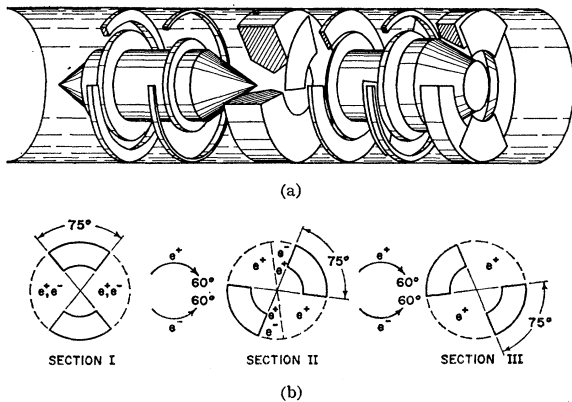


FIG. 6. (a) Two-cycle baffle of homogeneous field spectrometer, together with electron-positron discrimination baffles. (b) Three sections of electron-positron discrimination baffles.

The side wall of the counter was made out of 0.0015-in.-thick aluminum. A thin copper plating on the inside of this wall was necessary to avoid spurious pulses and to improve the plateau. The effective length of the counter was  $1\frac{3}{4}$  in. and its diam  $\frac{5}{8}$  in. A 0.003-in.-diam stainless-steel wire was used as an anode. The counter was filled with 1 atm of *Q*-gas (98.7% He, 1.3% butane). Under these conditions a plateau of 200–300 V was obtained. A device was constructed which allowed adjustment of the counter position from the outside of the spectrometer in three orthogonal directions. With this arrangement one can readily locate the optimum position of the counter which coincides with the line focus and thus gives maximum counting rate.

A hollow lead cylinder,  $4\frac{1}{2}$ -in. inner diam,  $8\frac{3}{8}$ -in. outer diam, and 4 in. long, covered both inside and outside with  $\frac{1}{8}$ -in.-thick aluminum, was made as shielding for the counter. With the arrangement described above the detector efficiency for focused electrons (and positrons) from a  $\frac{1}{8}$ -in.-diam source was 79%. Background was 7 counts/min.

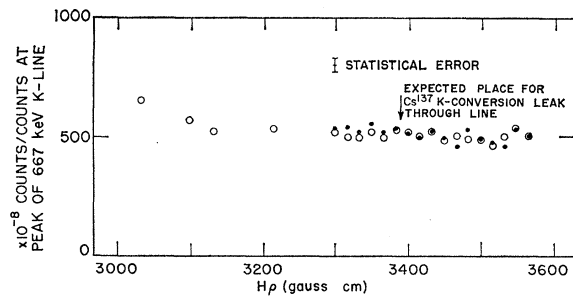


FIG. 7. Leak-through electron spectrum of  $\text{Cs}^{137}$  around 0.667-MeV *K* line.

Later during the experiment a commercial pancake counter was put into use. It had a diameter of  $1\frac{1}{4}$  in. and a detection efficiency of 50%. It recorded 11 background counts/min.

## MEASUREMENTS

The measurements started out with the 13-mC  $\text{Bi}^{206}$  source. The ring focus of the spectrometer was entirely open. In this case the spectrometer has a transmission of 0.65% (includes the transmission of the positron baffles) and a resolution of 2.2%. Preliminary measurements with sources from test bombardments had already shown that the positron spectrum of  $\text{Bi}^{206}$  does not end at 0.70 MeV, which is the end-point of the continuous distribution of positrons from the pairs belonging to the 1.72-MeV *E1* transition in  $\text{Pb}^{206}$ . This was unexpected from the decay scheme for  $\text{Bi}^{206}$ , as suggested by Alburger and Pryce.<sup>4</sup> In their decay scheme, no transition of an energy higher than 1.72 MeV was reported. The weak positron spectrum discovered above 0.70 MeV seemed to have an end-point around 0.90 MeV and, therefore, unfortunately covers the range

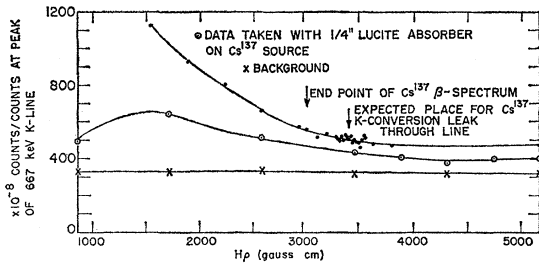


FIG. 8. Leak-through electron spectrum of  $\text{Cs}^{137}$  in the energy range 0.1–1.0 MeV.

where the positron line is expected (i.e., 0.79 MeV). The positrons observed between 0.70 and 0.90 MeV seemed to belong to the internal pairs of a 1.88-MeV transition decaying with the half-life of  $\text{Bi}^{206}$ . This was confirmed by observing the existence of its  $K$ - and  $L$ -conversion lines. A detailed measurement of the positron spectrum around 0.79 MeV (which corresponds to 3970 G cm) (Fig. 10) does not show any indication of a positron line within the limits of statistical uncertainty which is  $(N_{e_m^{+K}}/N_{e^{-K}})_{1.72 \text{ MeV}} \leq 5.5 \times 10^{-5}$  or using the  $K$ -conversion coefficient<sup>8,9</sup>  $\alpha_{eK} = 0.0008$ ,  $(N_{e_m^{+K}}/N_\gamma)_{1.72 \text{ MeV}} \leq 4.4 \times 10^{-8}$ .

More accurate results were obtained when the 80-mC  $\text{Bi}^{206}$  was used. The ring-focus slit of the spectrometer was now partly closed, which gave an improved resolution of 1.4%. The transmission of the spectrometer went down to 0.35%. In order to take advantage of the strong initial source the positron spectrum around 0.79 MeV was first investigated in greatest detail.  $\frac{1}{2}$ -h measurements were made for 20 values of current corresponding to momenta between 3800 and 4150 G cm. This was repeated 5 times, spectra with increasing values of momenta and decreasing ones being taken alternately. After this the immediate region around 3970 G cm was measured for another 4 h, the previously measured points being repeated and additional ones being put in between them. In every measurement, decay corrections were applied in such a way, that all the counting rates obtained in one spectrum were

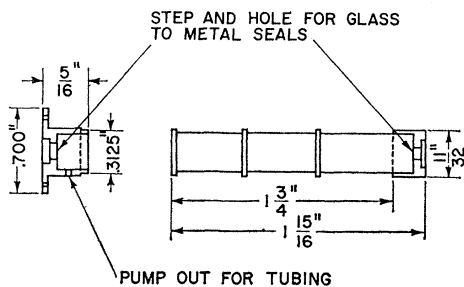


FIG. 9. Small side-window cylindrical Geiger counter.

<sup>8</sup> T. Novakov, S. Hultberg, and B. Andersson, *Arkiv Fysik* **13**, 117 (1958).

<sup>9</sup> R. Stockendal and S. Hultberg, *Arkiv Fysik* **15**, 33 (1959).

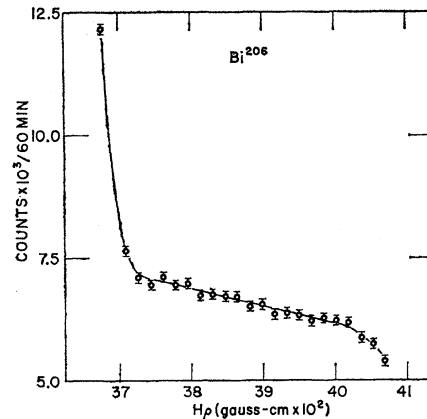


FIG. 10. Positron spectrum around 0.79 MeV of 13-mC  $\text{Bi}^{206}$  source.

normalized to the time at which the first point of the spectrum was taken. An empirical decay constant, obtained from the decay curves of several points in the energy region under consideration was used for the corrections. Figure 11 shows the added spectra where each point is normalized to a total counting time of  $2\frac{1}{2}$  h. Figure 12 shows the same spectrum after the counting rate of each point has been normalized by its momentum. From Figs. 12 and 13 one can say with fair certainty that it looks like there does exist a peak in the positron spectrum at the place where the line is expected (i.e., for  $H\rho = 3970$  G cm). Assuming that this peak can be attributed to the positron line, one gets  $(N_{e_m^{+K}}/N_{e^{-K}})_{1.72 \text{ MeV}} = 4.0 \times 10^{-5}$  or with<sup>8,9</sup>  $\alpha_{eK} = 0.0008$ ,  $(N_{e_m^{+K}}/N_\gamma)_{1.72 \text{ MeV}} = 3.2 \times 10^{-8}$ .

The positron spectrum in the entire region between 0.4 and 0.9 MeV was then carefully measured. It is shown in Fig. 13. The weak positron spectrum previously believed to belong to the internal pairs of a single electromagnetic transition of 1.88 MeV now turned out to be made up of three different parts, each part belonging to the internal pairs of three electromagnetic transitions of energies 1.845, 1.88, and 1.90 MeV, respectively. An accurate measurement of the electron spectrum showed the  $K$  and  $L$  conversion lines of these three different  $\gamma$ -ray transitions. This can be seen on Figs. 14(a) and (b). Figure 14(b) is obtained by subtracting the 1.88-MeV  $K$ -conversion

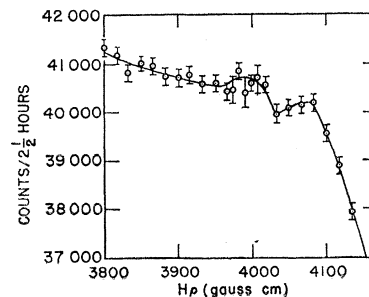


FIG. 11. Positron spectrum around 0.79 MeV of 80-mC  $\text{Bi}^{206}$  source.

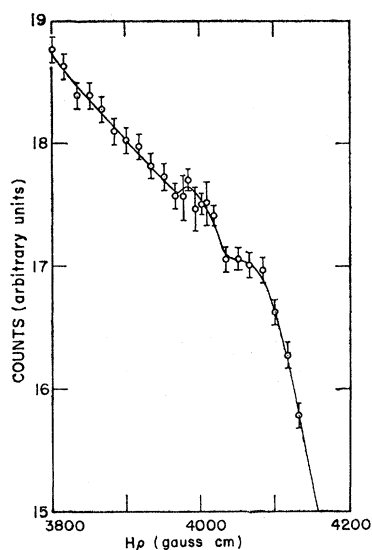


FIG. 12. Normalized positron spectrum around 0.79 MeV of 80-mC  $\text{Bi}^{206}$  source.

line with its proper resolution from the measured spectrum. The 1.845-MeV  $K$  line is only partly resolved from the 1.72-MeV  $L$  line, while the 1.90-MeV  $K$  line is broadened on its high-energy side because of the presence of the 1.845-MeV  $L$  line. Table III shows the

TABLE III. Intensities of the high-energy  $K$ -conversion lines in  $\text{Pb}^{206}$ .

$E$ (MeV)	$K$ -line intensity (electrons per 100 disintegrations)
1.72	0.029 <sup>a</sup>
1.845	0.00055
1.88	0.00160
1.90	0.00024

<sup>a</sup> See reference 4.

intensities of the three discovered electron lines as they are obtained from comparison with the 1.72-MeV  $K$  line.

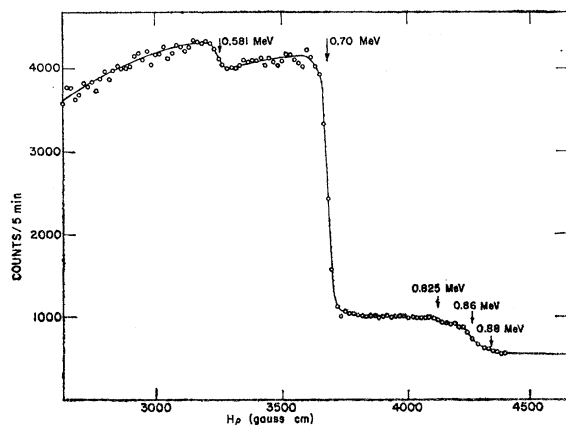


FIG. 13. Positron spectrum in the energy region 0.4-0.9 MeV of 80-mC  $\text{Bi}^{206}$  source.

TABLE IV. Assignment of multiplicities to the high-energy  $\gamma$ -ray transitions in  $\text{Pb}^{206}$  by comparing the experimental and theoretical values for  $\alpha_\pi/\alpha_K$  of each transition.

$E$ (MeV)	Exp.		Theor.		
	$\frac{\alpha_\pi}{\alpha_K}$	$\frac{\alpha_\pi}{\alpha_K}$ ( $E0$ )	$\frac{\alpha_\pi}{\alpha_K}$ ( $E1$ )	$\frac{\alpha_\pi}{\alpha_K}$ ( $E2$ )	$\frac{\alpha_\pi}{\alpha_K}$ ( $M1$ )
1.72	0.33	0.037	0.33	0.078	0.020
1.845	0.52	0.058	0.50	0.097	0.028
1.88	0.53	0.066	0.54	0.109	0.033
1.90	0.82	0.070	0.56	0.116	0.035

Information on the multiplicities of the 1.845, 1.88, and 1.90-MeV transitions can be obtained by comparing the measured ratios of internal pairs to  $K$ -conver-

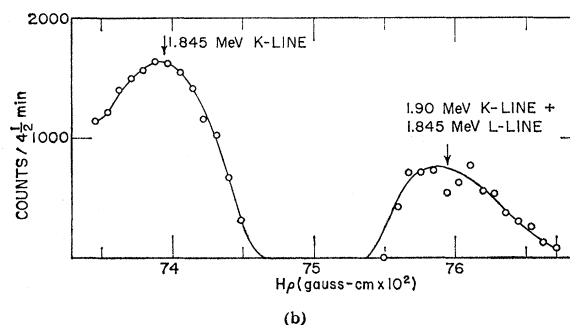
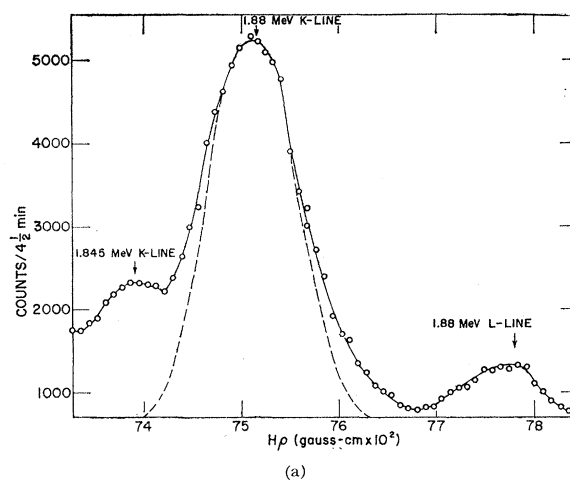


FIG. 14. (a) High-energy electron spectrum of  $\text{Bi}^{206}$ . (b) High-energy electron spectrum of  $\text{Bi}^{206}$  after subtraction of 1.88-MeV  $K$  line and background.

sion electrons. In order to get the total amount of internal pairs belonging to each separate transition the individual steps in the measured spectrum were extrapolated towards the region of low energy by assuming the theoretical energy distribution of positrons from internal pairs calculated by Jaeger and Hulme<sup>10</sup> for  $Z=83$ . Table IV shows the experimental values

<sup>10</sup> J. C. Jaeger and H. R. Hulme, Proc. Roy. Soc. (London) 148, 708 (1935).

TABLE V. Estimation of the retardation factor of the 1.72-MeV  $E1$  transition in  $Pb^{206}$  obtained by comparing the measured intensity of this transition and other  $M1$  transitions also depopulating the 3.403-MeV 5- level in  $Pb^{206}$  to the corresponding theoretical intensities predicted by the single-particle model.

$E$ (MeV)	$\left[ \frac{I_{1.72 \text{ MeV}}(E1)}{I_E(M1)} \right]_{\text{exp}}$	$\left[ \frac{I_{1.72 \text{ MeV}}(E1)}{I_E(M1)} \right]_{\text{s.p.m.}}$	$\left[ \frac{I_{1.72 \text{ MeV}}(E1)}{I_E(M1)} \right]_{\text{s.p.m.}} / \left[ \frac{I_{1.72 \text{ MeV}}(E1)}{I_E(M1)} \right]_{\text{exp}}$	$f_{\text{ret}}$
1.02	4.75 <sup>a</sup>	850	180	180-1800
0.62	5.7 <sup>a</sup>	3800	670	670-6700
0.39	57 <sup>a</sup>	16000	280	280-2800

<sup>a</sup> See reference 4.

obtained for the 1.72, 1.845, 1.88, and 1.90-MeV transitions. These values are compared to the theoretical ones corresponding to the multiplicities of  $E0, E1, E2$ , and  $M1$ .<sup>10,11-13</sup> As can be easily seen, the experimental values for  $\alpha_{\pi}/\alpha_{eK^-}$  are by far closest to the theoretical ones for  $E1$  transitions. Only for the 1.90-MeV transition the value of  $(\alpha_{\pi}/\alpha_{eK^-})_{\text{exp}}$  is considerably higher than the theoretical value for an  $E1$  transition. Since, however,  $(\alpha_{\pi}/\alpha_{eK^-})_{\text{theor}}$  is larger for  $E1$  than the corresponding value for any other multiplicity, there is some ground for the assumption that the 1.90-MeV transition as well as the other ones are of the  $E1$  type. Similar results on the intensities and multiplicities of the three high-energy electromagnetic transitions in  $Pb^{206}$  were also obtained by Brunner, Lombard, Perdrisat, and Leisi.<sup>14</sup>

### DISCUSSION

By comparing the upper limit  $(N_{e_m+K}/N_{\gamma})_{1.72 \text{ MeV}} \leq 4.4 \times 10^{-8}$ , which was obtained from measurements with the first source to the intensity  $(N_{e_m+K}/N_{\gamma})_{1.72 \text{ MeV}} = 3.2 \times 10^{-8}$  which was found for the peak at 0.79 MeV in the positron spectrum of the second source, it is obvious that these two results are consistent. There were two reasons why the peak in the positron spectrum of the first source was not observed:

1. The statistical uncertainty of the measurements was too large.

2. The  $Bi^{205}$  contamination (0.25%) was large enough to interfere seriously with the measurement of the positron line. This can be illustrated in the following way: The reduction in counting rate between consecutive points on the smooth curve of Fig. 11 is around 1%. This is in agreement with the calculated spectrum for a  $Bi^{206}$  source containing 0.25%  $Bi^{205}$ , shown in Fig. 15. The slope of the positron spectrum of the second source, where the  $Bi^{205}$  contamination is as small as 0.05%, is 4 times smaller than the one obtained for the first source, so the difference in counting rate of two consecutive points is only 0.25%. The peak which was found in the positron spectrum of the

second source is only 1% of the background (internal pairs from  $Bi^{206}$ ,  $\beta^+$  spectrum and internal pairs from  $Bi^{205}$ ) on which it is superimposed. In the case of the first source, peak/background should have been slightly smaller than 1% because of increased  $Bi^{205}$  background. It is, therefore, clear that even with improved statistics the detection of the peak in the positron spectrum of the first source would have been very difficult.

Perdrisat, Brunner, and Leisi<sup>15</sup> have recently repeated Brunner *et al.* measurements of the positron line in  $Bi^{206}$  and now obtain the value  $(N_{e_m+K}/N_{\gamma})_{1.72 \text{ MeV}} = (7.2 \pm 2.1) \times 10^{-8}$ . It might be interesting to point out that they used a source, where the  $Bi^{205}$  contamination was as high as 1%. The height of the peak is here about three times smaller than the difference in counting rate of consecutive points of the underlying background.

The question now arises if the peak which was found in the positron spectrum of the second source really can be attributed to a monoenergetic positron line with the intensity  $N_{e_m+K}/N_{\gamma} = 3.2 \times 10^{-8}$ . It can be explained in two additional ways of which at least the first one seems very unlikely:

1. The  $K$ -conversion electron lines of the 0.880-MeV and 0.895-MeV transitions in  $Pb^{206}$  have the energies 0.792 and 0.808 MeV, respectively. Those could possibly give a common or two separate "leak-through" lines at the energy at which the positron peak was observed. Taking into account the measured limit for

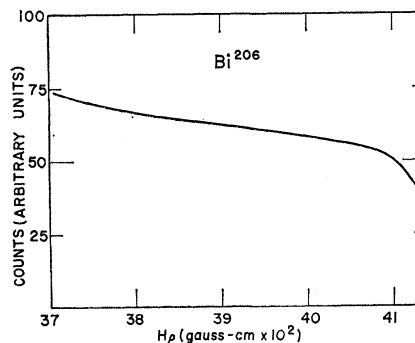


FIG. 15. Calculated positron spectrum of  $Bi^{206}$  with 0.25%  $Bi^{205}$  contamination.

<sup>11</sup> M. H. Wang, *Nature* **162**, 264 (1948).

<sup>12</sup> M. E. Rose, G. H. Goertzel, B. I. Spinrad, J. Harr, and P. Strong, *Phys. Rev.* **83**, 79 (1951).

<sup>13</sup> R. Thomas, *Phys. Rev.* **58**, 714 (1940).

<sup>14</sup> H. Brunner, R. Lombard, C. F. Perdrisat, and H. T. Leisi, *Helv. Phys. Acta* **34**, 472 (1961).

<sup>15</sup> C. F. Perdrisat, J. H. Brunner, and H. J. Leisi, *Helv. Phys. Acta*, **35**, 175 (1962).



the leak-through factor ( $10^{-7}$ ) and the strength of these electron lines relative to the 1.72-MeV  $K$  line one gets

$$\frac{[(N_{e^{-K}})_{0.880 \text{ MeV}} + (N_{e^{-K}})_{0.895 \text{ MeV}}]_{\text{leak through}}}{(N_{e^{-K}})_{1.72 \text{ MeV}}} \leq 2.8 \times 10^{-6}.$$

This is more than ten times smaller than the measured value  $(N_{e_m^{+K}}/N_{e^{-K}})_{1.72 \text{ MeV}} = 4 \times 10^{-5}$  which led to  $(N_{e_m^{+K}}/N_{\gamma})_{1.72 \text{ MeV}} = 3.2 \times 10^{-8}$ .

2. There could exist an additional electromagnetic transition with an energy around 1.79 MeV. This transition would then have internal pairs of which the edge of the positron spectrum would fall in the place of the spectrum where the positron peak was observed. Unless this transition is of the  $0^- - 0^+$  type it should be accompanied by its electron conversion lines. Unfortunately, the  $K$ -conversion line would almost coincide with the relatively strong  $L$ -conversion line of the 1.72-MeV transition and might not have been resolved in the electron spectrum of  $\text{Bi}^{206}$ .

The discrepancy between the theoretical and the experimental values of  $\alpha_{\pi}/\alpha_{e_K}$  - which was found in the case of the 1.90-MeV transition can be explained in several ways. It was difficult to estimate the height of its  $K$ -conversion line because of its low intensity and the presence of the neighboring 1.845-MeV  $L$ -conversion line. Likewise there exists some uncertainty in the evaluation of the intensity of the internal pairs because of the unknown shape of the background on which the edge of the positron spectrum is superimposed. This background does not necessarily have to be constant with current since it may contain leak through electrons and  $\beta^+$  particles from the source.

Furthermore because of the unknown shape of the leak-through electron spectrum from the source it is impossible to say whether there exists a very weak  $\beta^+$  transition in  $\text{Bi}^{206}$ . Such a transition has been reported by Perdrisat *et al.*<sup>15</sup>

### CONCLUSION

As was pointed out in the Introduction, the knowledge of the intensity of the monoenergetic positron line of a certain multipole transition gives an estimate for the lifetime of the level from which the transition originates. For the 1.72-MeV  $E1$  transition in  $\text{Pb}^{206}$  the different constants in formula (1) have the following values:

$\alpha_{e_m^{+K}} = 8.25 \times 10^{-5}$  according to Lombard and Rys.<sup>2</sup>  $P_{\gamma}^{+K} = 0.72$ , assuming an energy difference of 0.30-MeV between the ground state of  $\text{Bi}^{206}$  and the 3.403-MeV level of  $\text{Pb}^{206}$  and that the  $\beta$  transition to the 3.403-level is first forbidden.<sup>16-18</sup>  $\tau_K = 1.15 \times 10^{-17}$  sec.<sup>17</sup> One then gets for the lifetime of the 3.403-MeV level in  $\text{Pb}^{206}$   $\tau_n = 2.1 \times 10^{-14}$  sec. If one compares  $\tau_n = 2.1 \times 10^{-14}$  sec to the corresponding value expected from the single-particle model,  $\tau_n^{s.p.m.} = 3.6 \times 10^{-17}$  sec, then the retardation factor  $f_{\text{ret}} = \tau_n / \tau_n^{s.p.m.} \sim 600$  is obtained.

Some idea about whether a retardation factor of 600 makes sense or not in this case can be obtained by comparing the ratio of the measured intensity of the 1.72-MeV  $E1$  transition to those of the competing  $M1$  transitions. The conversion coefficients of the latter transitions have been measured quite accurately<sup>8,9</sup> and as a result from those one can conclude that their  $E2$  admixture is very small. They should, therefore, not be retarded very much themselves and it should be fair to assume that their retardation factors are between 1 and 10. Table V gives an estimate of the retardation factor for the 1.72-MeV  $E1$  transition from the comparison of the intensity of this transition to those of three out of four  $M1$  transitions depopulating the 3.403-MeV 5- level in  $\text{Pb}^{206}$ .

The measured retardation factor 600 seems to lie in the range of corresponding values obtained from Table V.

The three high-energy transitions in  $\text{Pb}^{206}$ , reported in this work, do not fit into the decay scheme of  $\text{Bi}^{206}$ , given by Alburger and Pryce.<sup>4</sup> Coincidence measurements are necessary to show their origins.

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<sup>16</sup> H. Brysk and M. E. Rose, *Rev. Mod. Phys.* **30**, 1169 (1958).

<sup>17</sup> M. Mladjenović, *Arkiv Fysik* **8**, 27 (1954).