

wave measurements<sup>3</sup> by the same authors have given a ratio in much closer agreement. The crystal structure of  $\text{SnI}_4$  is known,<sup>4</sup> and Dehmelt<sup>5</sup> has made and discussed higher temperature measurements of the  $\text{I}^{127}$  resonance. The lattice is cubic with two nonequivalent iodine lattice positions. One-quarter of the iodine atoms lie on  $C_3$  axes of the unit cube and this symmetry requires  $\eta=0$  for these atoms. The crystal symmetry does not require  $\eta=0$  for the remaining three quarters of the

<sup>3</sup> R. Livingston *et al.* (unpublished results).

<sup>4</sup> R. W. G. Wyckoff, *Crystal Structures* (Interscience Publishers, New York, 1951).

<sup>5</sup> H. G. Dehmelt, *Z. Physik* **130**, 356 (1951).

iodine atoms. The type *B* lines for which  $\eta=0$  are appropriately weaker than the corresponding type *A* lines in agreement with the crystal structure. Dehmelt's values for  $\eta$ , determined for  $\text{I}^{127}$  at room temperature, are the same as the lower temperature values for  $\text{I}^{129}$  given above. His quadrupole coupling value for  $\text{I}^{127}$  is appropriately lower than the above values because of the temperature difference.

We wish to acknowledge the help of Gordon Hebert and George Parker of the Chemistry Division, this Laboratory, for supplying the  $\text{I}^{129}$  which they isolated from fission material.

## Comparison of the Values of the Disintegration Constant of $\text{Be}^7$ in Be, BeO, and $\text{BeF}_2$ \*

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The difference in the decay constants of  $\text{Be}^7$  in Be, BeO, and  $\text{BeF}_2$ , have been measured using the differential ionization chamber technique. The results are  $\lambda(\text{Be}) - \lambda(\text{BeF}_2) = (7.41 \pm 0.47) \times 10^{-4} \lambda$ ,  $\lambda(\text{BeO}) - \lambda(\text{BeF}_2) = (6.09 \pm 0.55) \times 10^{-4} \lambda$ ,  $\lambda(\text{Be}) - \lambda(\text{BeO}) = (1.31 \pm 0.51) \times 10^{-4} \lambda$ . These values are smaller than the results previously obtained by Leininger, Segrè, and Wiegand for the same compounds, but support their earlier work.

THE possibility of altering the decay rate of  $\text{Be}^7$  by changing the electron density at the nucleus was suggested by Segrè<sup>1</sup> and by Daudel.<sup>2</sup> Experiments performed to determine the influence of different states of chemical combination of beryllium on the decay constant of  $\text{Be}^7$  have given the results listed in Table I.

The present series of measurements was undertaken in order to compare  $\lambda(\text{Be})$ ,  $\lambda(\text{BeO})$ , and  $\lambda(\text{BeF}_2)$  in the same experiment, using the improved methods now available for the measurement of small currents and for the detection of source contaminants. The ionization currents were measured by a vibrating reed electrometer<sup>3</sup> used essentially as a null indicator, thus eliminating the effects of any variations in the gain or calibration of the instrument. The use of a scintillation counter,<sup>4,5</sup> not available in the earlier experiments,<sup>6</sup> is an aid in detecting and identifying radioactive contaminants.

$\lambda$ -values for different beryllium compounds were compared by the balanced ionization chamber method introduced in 1911 by Rutherford.<sup>7</sup> The particular ionization chambers and current measuring equipment for this experiment were those used by Bainbridge, Goldhaber, and Wilson<sup>8</sup> in the study of  $\text{Tc}^{99m}$ .

### I. EXPERIMENTAL PROCEDURE

#### A. Preparation of the Sources

The  $\text{Be}^7$  was obtained from a proton bombardment of cp metallic lithium with the 8.5-Mev proton beam from the cyclotron at the University of Pittsburgh. A radioautograph was taken of the bombarded target and the surface containing the  $\text{Be}^7$  activity was scraped and dissolved in dilute hydrochloric acid. The solution was made 0.3*N* in hydrochloric acid, and 50 milligrams each of iron, nickel, cobalt, copper, zinc, and cadmium were added. The main contaminant, copper, was precipitated by hydrogen sulfide. The hydrogen sulfide was removed from the supernatant, and the active  $\text{Be}^7$  was carried with  $\text{Fe}(\text{OH})_3$  precipitated by the addition of ammonium hydroxide. The  $\text{Fe}(\text{OH})_3$  was dissolved in dilute hydrochloric acid and again precipitated, in the presence of the hold back carriers, by ammonium

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<sup>1</sup> E. Segrè, *Phys. Rev.* **71**, 274 (1947).

<sup>2</sup> R. Daudel, *Rev. sci.* **85**, 162 (1947).

<sup>3</sup> Palevsky, Swank, and Grenchik, *Rev. Sci. Instr.* **18**, 298 (1947).

<sup>4</sup> H. Kallman, *Phys. Rev.* **75**, 623 (1949).

<sup>5</sup> R. Hofstadter, *Phys. Rev.* **74**, 100, 628 (1949).

<sup>6</sup> E. Segrè and C. E. Wiegand, *Phys. Rev.* **81**, 284 (1951); **75**, 39 (1949).

<sup>7</sup> E. Rutherford, *Wien. Ber.* **120**, 303 (1911).

<sup>8</sup> Bainbridge, Goldhaber, and Wilson, *Phys. Rev.* **90**, 430 (1953) [hereafter referred to as BGW].

hydroxide. This process was repeated three times, carefully washing the Fe(OH)<sub>3</sub> each time.

The Fe(OH)<sub>3</sub> solution was made 7.8*N* in HCl, and four iron extractions were made with isopropyl ether. The ether was evaporated from the aqueous phase and 62 milligrams of beryllium in the form of Be(NO<sub>3</sub>)<sub>2</sub> · 3H<sub>2</sub>O were added. On addition of ammonium hydroxide, Be(OH)<sub>2</sub> was precipitated and then washed three times with water. The Be(OH)<sub>2</sub> was dissolved in glacial acetic acid and taken to dryness several times in order to convert the hydroxide completely to the basic beryllium acetate. The basic acetate was extracted into chloroform and the chloroform layer thoroughly washed with water. The acetate was then taken to dryness and taken up with 10 cc of HNO<sub>3</sub>. This compound was again taken nearly to dryness and diluted with water.

The purity of the Be<sup>7</sup> was determined from an aliquot of this solution. Zinc and copper carriers were added to 1 percent of the total solution and precipitated with hydrogen sulfide at a *pH* of 2-3. The precipitate was washed, dried, and saved for analysis. Two precipitations of Be(OH)<sub>2</sub> were then made from the same aliquot in the presence of the same hold back carriers, and the filtrate from this precipitation was dried and combined with the zinc and copper residues. The gamma-ray spectrum of these residues was analyzed by Mr. Michael McKeown with a NaI scintillation spectrometer. The only photoelectric peak observed was that of the 0.48-Mev gamma-ray associated with Be<sup>7</sup>. The activity in the residues was 7 × 10<sup>-2</sup> percent of the total Be<sup>7</sup> activity in that aliquot. Any contaminants emitting gamma-rays of energies differing by 0.06 or more Mev from the 0.48-Mev radiation of Be<sup>7</sup> could have been detected if present to one percent or greater of the Be<sup>7</sup> activity in the residues. Gamma-ray contaminants therefore did not exceed the negligible amount of 7 × 10<sup>-6</sup> of the source activities. Another aliquot of 0.01 percent of the Be<sup>7</sup> solution was analyzed in the same way, and again only the peak associated with the 0.48 gamma-ray was observed.

The four sources used in the following measurements were then prepared from the remaining solution. Two identical sources of BeO were made by the ignition of Be(OH)<sub>2</sub> in platinum crucibles at 1000°C. The sources were housed in small brass capsules which had been lined with a thin layer of pure tin. Screw caps were then forced down on the oxides so as to prevent any mechanical movement during the measurements.

A beryllium fluoride source was prepared by the hydrofluorination of beryllium hydroxide. The hydroxide was held in a covered platinum boat and heated in a monel furnace for sixteen hours at a temperature of 200°C. The temperature was then raised to 500°C for five hours to sinter the beryllium fluoride. An x-ray analysis was performed by Mr. Otto F. Kammerer to determine the crystal structure of this fluoride and also of an earlier inactive sample of beryllium fluoride. The structure was found to be hexagonal with constants

TABLE I. Relative differences,  $\Delta\lambda/\lambda$ , in the decay constant  $\lambda$  of Be<sup>7</sup> in different chemical compounds. Errors are standard deviations.

Item	Comparison	10 <sup>4</sup> Δλ/λ	References
1	λ(Be) - λ(BeCl <sub>2</sub> ) BeCl <sub>2</sub> in solution	50 ± ~30	a, b
2	λ(Be) - λ(BeF <sub>2</sub> ) BeF <sub>2</sub> "noncrystalline"	100 ± 30	b, c, d
3	λ(Be) - λ(BeO)	0.69 ± 1.25 2.38 ± 1.5	e
4	Weighted mean λ(BeO) - λ(BeF <sub>2</sub> )	1.5 ± 0.9 6.9 ± 0.3	f
5 (3 plus 4)	λ(Be) - λ(BeF <sub>2</sub> )	8.4 ± 1	f

<sup>a</sup> Bouchez, Daudel, Daudel, and Muxart, *J. phys. et radium* **8**, 336 (1947).  
<sup>b</sup> Benoist, Bouchez, Daudel, Daudel, and Rogozinski, *Phys. Rev.* **76**, 1000 (1949).

<sup>c</sup> Bouchez, Daudel, Daudel, and Muxart, *Compt. rend.* **227**, 525 (1948).  
<sup>d</sup> Bouchez, Daudel, Daudel, Muxart, and Rogozinski, *J. phys. et radium* **10**, 201 (1949).

<sup>e</sup> E. Segrè and C. E. Wiegand, *Phys. Rev.* **81**, 284 (1951); **75**, 39 (1949).  
<sup>f</sup> Leininger, Segrè, and Wiegand, *Phys. Rev.* **81**, 280 (1951); **76**, 897 (1949).

$a = 4.72A$ ,  $c = 5.18A$ . This result is in agreement with the observations by Novoselava *et al.*<sup>9</sup> and later by Leininger, Segrè, and Wiegand.<sup>10</sup> The fluoride was housed in a small brass capsule similar to those used for the oxides, with Apiezon wax packed around the source to prevent motion within the housing and also to prevent any moisture from coming in contact with the fluoride.

The metal was prepared by an elegant method developed by Dr. T. T. Magel of Massachusetts Institute of Technology.<sup>11</sup> A mixture of beryllium oxide and zirconium powder was heated by induction in a tantalum crucible under vacuum. At 1300°C the reduced beryllium metal distilled away from the zirconium and was collected on a cone shaped cold finger directly above the crucible. The metal was housed in another brass capsule and protected from motion and moisture with Apiezon wax. The four sources were then matched to within three divisions in 7000 by machining the capsule walls of the weaker sources and by adding absorber to the walls of the stronger sources.

## B. Measurements of $\Delta\lambda/\lambda$

The apparatus and general technique used for the determination of  $\Delta\lambda/\lambda$  for sources in different chemical states of combination have been described at length in BGW. Inasmuch as the same equipment and essentially the same method were used in this work, only points of difference between the two experiments will be discussed.

Since the balanced ionization chambers provide a means for measuring the relative strength of two sources, there are six possible combinations of the four

<sup>9</sup> Novoselava, Levina, Simanov, and Zhasmin, *J. Gen. Chem (U.S.S.R.)* **14**, 385-402 (1944).

<sup>10</sup> Leininger, Segrè, and Wiegand, *Phys. Rev.* **81**, 280 (1951); **76**, 897 (1949).

<sup>11</sup> We are indebted to Dr. A. R. Kaufmann of the same institution for suggesting the use of Dr. Magel's method.

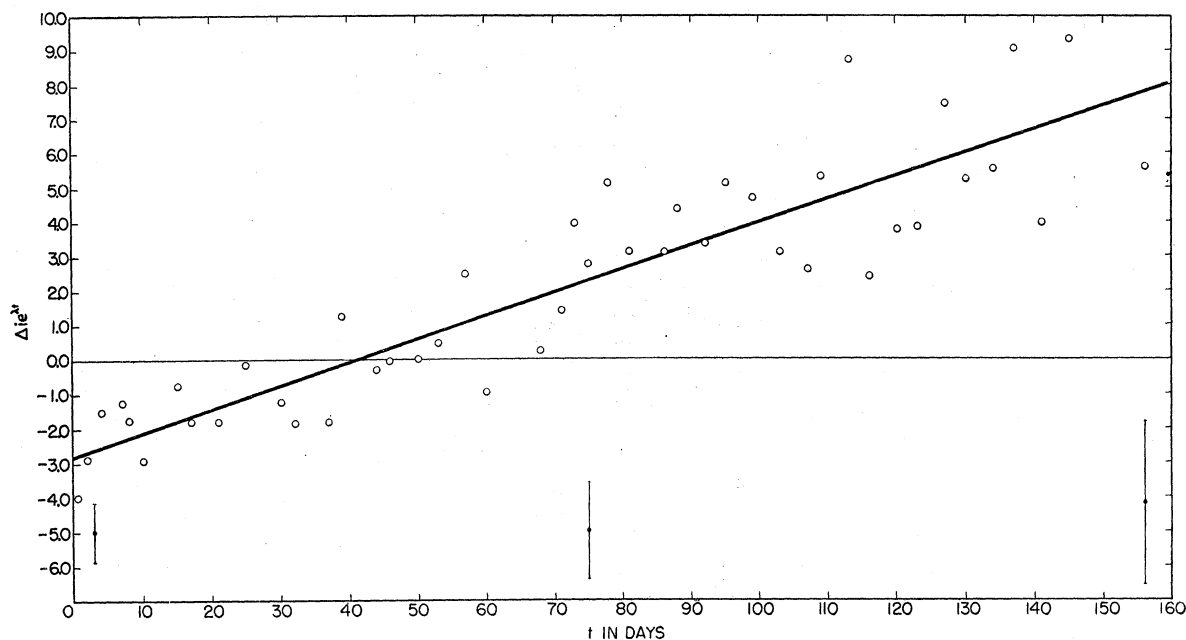


FIG. 1.  $e^{\lambda t} \Delta i$  versus time for the  $\text{BeF}_2$ -Be source set. The errors to be associated with the individual points are indicated at three representative times and are standard deviations based on external consistency.

sources described above that can be measured. Data from all six sets of sources can be the basis of imposing constraints in the analysis of the final results, leading to more accurate values of  $\Delta\lambda/\lambda$ . Further, comparison of the two oxide sources provides a means for observing instrumental effects and possible physical change in the sources.

The experiment consisted then in the determination of the difference current,  $\Delta i$ , as a function of time for each of the six sets of sources.  $\Delta i$  was measured by allowing the charge to accumulate for 1200 seconds on the plates of the quartz-insulated, vacuum dielectric,  $10^{-9}$  farad condenser. At the end of the 1200-second period the deflection as indicated on the chart of the recording potentiometer was matched by displacing the zero line. This displacement was produced by inserting a known voltage into the input of the vibrating reed electrometer by means of the potentiometer (Fig. 3, BGW).<sup>8</sup> The relationship  $\Delta i = c\Delta v/\Delta t$  was then used to calculate  $\Delta i$ .

For each set of sources a 1200 second run was made with each of the two sources in a particular chamber. Immediately following the first run the sources were interchanged and the run repeated. A value of  $\Delta i$  was obtained by taking an average for the two runs.

The potential on the chambers was maintained by a pack of 45 volt batteries, with a provision for small adjustments. At the beginning of a run on a set of sources, the potentials on the two chambers were adjusted to a pre-selected value to within 1 part in 10 000. If after a 1200 second run they were found to have drifted as much as 5 parts in 10 000, they were readjusted. Since in any case the two battery packs stay

close together as they drift, any induction effects cancelled and no error was introduced.

The sources were positioned in the chambers in a reproducible fashion as described in BGW.<sup>8</sup> The six sets of sources were measured using a routine that minimized their handling, and the sequence of operations remained the same for the length of the experiment. Measurements were taken twice a week over a five-month period (two mean-lives).

The currents were measured in units of  $10^{-14}$  ampere. In order to present the data in a linear form, the values of  $\Delta i$  were multiplied by  $e^{\lambda t}$ , as was done by Segrè and Wiegand<sup>6</sup> and BGW. The data from the sets of sources that were used in obtaining the final results are shown in Figs. 1, 2, and 3. The solid lines are those calculated by a least squares procedure from the data.

In addition to the data obtained for the difference current as a function of time, other measurements were made to monitor the performance of the instrument and to obtain a value of the decay constant for  $\text{Be}^7$ . As in the case of BGW, the ionization current was measured with a weak radium source (about  $5\mu\text{c}$ ) in one chamber and a blank source in the other. This measurement was made about once a week using two resistors of  $10^{10}$  ohms, and the rms deviation from the average value was found to be 0.50 percent for one resistor and 0.76 percent for the other. Less frequently this current was measured using the  $10^{-9}$  farad quartz-insulated condenser. In this way the values of  $R_1$  and  $R_2$  could be compared directly with the capacity of  $C$ .

The total ionization current at  $t=0$ ,  $I_0$ , which enters directly into the calculation of  $\Delta\lambda/\lambda$  from the slope of

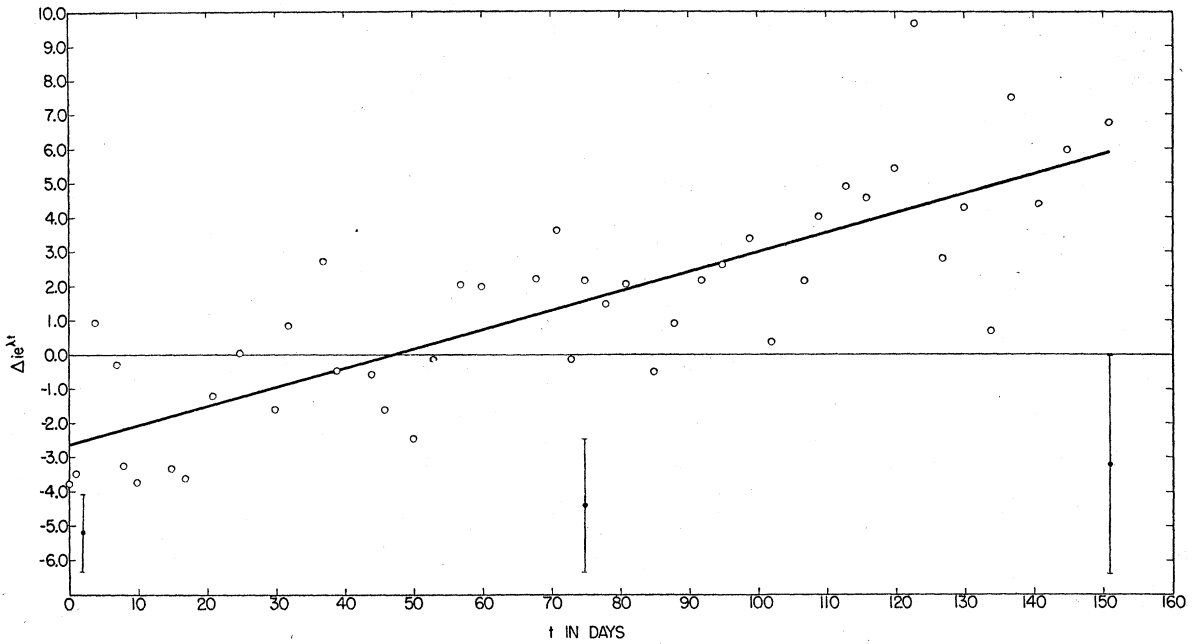


FIG. 2.  $e^{\lambda t} \Delta i$  versus time for the  $\text{BeF}_2$ - $\text{BeO}(1)$  source set.

the  $e^{\lambda t} \Delta i$  plot, was determined from a series of measurements extending over a period of five months, using  $R_1$  and  $R_2$ . These data from the metal source gave  $\lambda$  and  $I_0$  for  $\text{Be}^7$ . By the comparison of  $R_1$ ,  $R_2$  and  $C$  with the weak radium source, the intensity  $I_0$  could be expressed directly in terms of  $C$ , which was used for the  $\Delta i$  determinations.

Since  $R_1$ ,  $R_2$ , and  $C$  were compared at about 480 divisions, and  $I_0$  was 7133 divisions, it was important to establish the presence of any voltage coefficient of re-

sistance for  $R_1$  and  $R_2$ . Several times during the course of the experiment, a resistor calibration was carried out in the fashion described in BGW, and no measurable coefficient was found.

## II. EXPERIMENTAL RESULTS

### A. Determination of $\lambda$

The data on the direct measurement of the metal source with  $R_1$  and  $R_2$  were analyzed to determine the

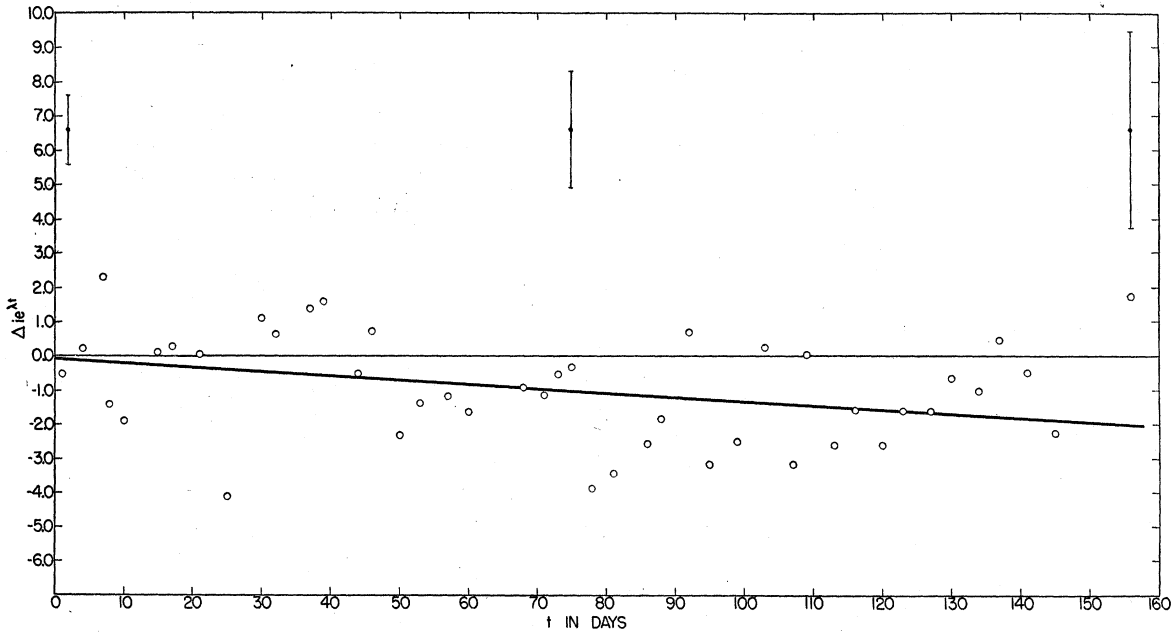


FIG. 3.  $e^{\lambda t} \Delta i$  versus time for the  $\text{Be}$ - $\text{BeO}(1)$  source set.

TABLE II. Results of the least squares analysis of the data for  $e^{\lambda t} \Delta i$  versus  $t$ . The errors indicated are standard deviations based on external consistency.

No.	Sources	Intercept	Slope
1.	BeF <sub>2</sub> -Be	-2.81±0.29	0.0678±0.0047
2.	BeF <sub>2</sub> -BeO(1)	-2.67±0.40	0.0568±0.0064
3.	Be-BeO(1)	-0.08±0.37	-0.0125±0.0057
4.	BeO(1)-BeO(2)	1.30±0.31	-0.0035±0.0049
5.	Be-BeO(2)	0.78±0.41	-0.0114±0.0065
6.	BeF <sub>2</sub> -BeO(2)	-2.26±0.33	0.0630±0.0053

decay constant. An analysis of the data using  $\ln I = \ln I_0 - \lambda t$  as a basis for the least squares procedure gave  $\lambda = 1.293 \pm 0.004 \times 10^{-2} \text{ day}^{-1}$ . This result includes a correction of one part in 350 to take account of the effect of departure of the chamber characteristics from ideally complete saturation. The half-life is  $53.61 \pm 0.17$  days.

Previous determinations of the half-life of Be<sup>7</sup> have been made by Hill,<sup>12</sup>  $53 \pm 2$  days; by Bouchez *et al.*,<sup>13</sup>  $54.3 \pm 0.5$  days; by Bonner,<sup>14</sup>  $54.5$  days; and by Segrè and Wiegand,<sup>6</sup>  $52.93 \pm 0.22$  days.

### B. Determination of $\Delta\lambda/\lambda$

The results of the least squares analysis of the data for the six sets of sources are shown in Table II. It can be seen that the comparison of the two oxide sources yielded no slope within the experimental error. Plots of the BeF<sub>2</sub>-BeO(2) measurements and others involving BeO(2) indicated a change of slope at about 110 days. This was interpreted as a possible shift in the position of oxide (2) within the capsule. For this reason the data involving BeO(2) were not utilized in computing the final results. It should be noted that the slopes of the BeO(1) plots are not inconsistent with the BeO(2) plots, and that if all of the data were included the final results would not be altered beyond the experimental errors.

<sup>12</sup> J. E. Hill, *Phys. Rev.* **57**, 567 (1940).

<sup>13</sup> Bouchez, Daudel, Daudel, and Muxart, *J. phys. et radium* **8**, 336 (1947).

<sup>14</sup> J. F. Bonner, Jr., Atomic Energy Commission Report AECU-107, unpublished.

The slope  $b$  is related to the desired quantity,  $\Delta\lambda/\lambda$ , by the relationship  $\Delta\lambda/\lambda = b/I_0\lambda$ . As previously stated, values of  $\lambda$  and  $I_0$  were obtained by a measurement of the intensity of the metal source as a function of time. In terms of the capacity  $C$  of  $10^{-9}$  farad,  $I_0$  was found to be 7133 divisions ( $10^{-14}$  ampere per division). Values of  $\Delta\lambda/\lambda$  have been calculated then for the three sets of sources involved and are shown in Table III.

The relationship that exists between the values of  $\Delta\lambda/\lambda$  for the three source comparisons can be utilized to yield a more accurate knowledge of each of these values. A constraint fitting using a least squares procedure that followed the method of Deming<sup>15</sup> has been employed, and the new values of  $\Delta\lambda/\lambda$  obtained as a result of this constraint fitting are also shown in Table III.

TABLE III. Relative differences in the decay constant of Be<sup>7</sup> in beryllium metal, oxide, and fluoride.

Sources	Values from data of Table II	Values adjusted by a constraint fitting
$\frac{\lambda(\text{Be}) - \lambda(\text{BeF}_2)}{\lambda} \times 10^4$	7.37±0.53	7.41±0.47
$\frac{\lambda(\text{BeO}) - \lambda(\text{BeF}_2)}{\lambda} \times 10^4$	6.15±0.73	6.09±0.55
$\frac{\lambda(\text{Be}) - \lambda(\text{BeO})}{\lambda} \times 10^4$	1.36±0.63	1.31±0.51

Our values of  $\Delta\lambda/\lambda$  for (BeO-BeF<sub>2</sub>) and (Be-BeF<sub>2</sub>) are lower, by about twice the standard deviations, than those obtained by Leininger, Segrè and Wiegand,<sup>6,10</sup> but essentially these results confirm their earlier work.

The authors take pleasure in thanking Miss Jean Snover for carrying out the extensive calculations in this work. We also wish to thank J. A. Miskel for help in the preparation of the beryllium sources and R. Davis for aid in the preparation of the cyclotron target. The information contained in the notes of L. K. Nash and E. L. King was extremely useful in the purification of the Be<sup>7</sup>.

<sup>15</sup> W. E. Deming, *Statistical Adjustment of Data* (John Wiley and Sons, Inc., New York, 1946).