ever the remaining discrepancies indicate that this model requires further development.

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Alteration of the Decay Constant of Te^{125m} by Chemical Means*

A. C. MALLIARIS AND KENNETH T. BAINBRIDGE
Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts

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The total rate of a nuclear transition which proceeds partially by internal conversion may be expected to change when alterations are produced in the states of the electrons concerned. Such alterations may be produced, for instance, by chemical means. Differences in the decay constant of the highly converted isomeric transitions of Te^{125m} have been measured using Te, TeO_2, and Ag_2Te (synthetic hessite). The results are: $\lambda(\mathrm{Te})-\lambda(\mathrm{Ag_2Te})=(2.59\pm0.18)\times10^{-4}\lambda(\mathrm{Te})$; $\lambda(\mathrm{TeO_2})-\lambda(\mathrm{Ag_2Te})=(2.23\pm0.18)\times10^{-4}\lambda(\mathrm{Te})$; $\lambda(\mathrm{Te})-\lambda(\mathrm{TeO_2})=(0.36\pm0.17)\times10^{-4}\lambda(\mathrm{Te})$. The errors are standard deviations arising mainly from the statistical fluctuations of the number of ionizing events which contributed to a measurement by the double-ionization-chamber balance method. The ionization currents were produced by the K x rays following the internal conversion process in the sources. Corrections were made for the effect of an initial 0.05% contamination of Te^{123m}. Sources of errors are considered in detail.

I. INTRODUCTION

THE decay constant of an isomer may be changed by external means if the outer electrons participate appreciably in the transition.[1] For example, a change of the chemical environment of the atoms concerned may cause a change of the electron density in the appropriate domain, thus speeding or slowing the nuclear transition.[2] Alterations in the decay rate have been produced also by applied high pressure,[3] as well as by bringing the element concerned into its superconducting state.[4]

This paper presents detailed results showing that the decay rate of the highly converted isomeric transition in Te^{125m} depends upon the chemical state of combination of the tellurium.[5] The decay constants were compared for Te^{125m} in Te (elemental), TeO_2, and Ag_2Te (synthetic hessite).

The decay scheme of Te^{125m} is given in Fig. 1.[6] The half-life is approximately 58 days. Since, as shown in Fig. 1, the outer electrons participate in the internal conversion process and there is no crossover transition, it appeared that the transition rate might be altered by chemical means. Another factor in the selection of Te^{125m} for these experiments was that tellurium under pressure undergoes a polymorphic transition and changes from a poor to a metallic conductor.[7] If Te^{125m} responded to the chemical influence, such a result would encourage extension of the experiments to later study of the possible effects of the interesting changes in lattice structure and electrical conductivity induced by pressure.

The decay rates of the different sources were compared by the differential ionization chamber method described in Ref. 2, but the steel chambers were spherical

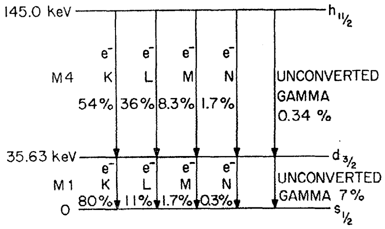

FIG. 1. The decay scheme of Te^{125m}.

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* Research supported in part by the Navy Department (Office of Naval Research) under ONR Contract Nonr-1866(19).

[1] R. Daudel, Rev. Sci. **85**, 162 (1947); and M. Goldhaber (unpublished lectures).

[2] K. T. Bainbridge, M. Goldhaber, and E. Wilson, Phys. Rev. **84**, 1260 (1951); **90**, 430 (1953); J. A, Cooper, J. M. Hollander, and J. O. Rasmussen, Phys. Rev. Letters **15**, 680 (1965); S. Shimizu and H. Mazaki, Phys. Letters **17**, 275 (1965).

[3] K. T. Bainbridge, Chem. Eng. News **30**, 654 (1952); R. A. Porter and W. G. McMillan, Phys. Rev. **117**, 795 (1960).

[4] D. H. Byers and R. Stump, Phys. Rev. **112**, 77 (1958).

[5] M. Goldhaber (private communication) mentioned Te^{125m} as an isomer which might show an effect.

[6] R. Narcisi, thesis, Harvard University, 1959 (unpublished).

[7] P. W. Bridgman, Proc. Am. Acad. Arts Sci. **81**, 167 (1952).

in shape with Dural thimbles to enclose the sources (25% absorption of the 27-keV radiation) instead of steel thimbles (75% absorption).

The present measurements were complicated by the presence of approximately 0.05% activity of Te^{123m} relative to the initial activity of the Te^{125m}. The Te^{123m} was formed by a series of (n,γ) reactions, starting with Sn^{120}, and by product decays which were completed in a time short compared with the irradiation time of the natural tin. The desired source Te^{125m} was produced from the standard sequence $Sn^{124}(n,\gamma)Sn^{125} \rightarrow Sb^{125} \rightarrow Te^{125m}$. In prior experiments on Te^{99m}, the only contaminant Re^{186} could be kept below 3×10^{-6} of the initial strength of the source, and its effects were negligible.[2] In the present experiments the measurements and corrections for the contribution of the contaminant will be discussed in detail.

Five sources in all: two Te, two TeO_2, and one Ag_2Te, were compared to give ten sets of difference measurements. From these a constraint fitting was finally obtained to yield the values of $\Delta\lambda/\lambda$ between Te, TeO_2, and Ag_2Te. The sources of error are discussed.

II. METHOD OF MEASUREMENT

The small differences in λ occuring between two sources were measured by the differential ionization chamber method first used by Rutherford for a similar purpose.[8] The equipment used here comprised two spherical steel ionization chambers with re-entrant Dural thimbles in which the sources (1) decay constant λ, and (2) decay constant $\lambda+\Delta\lambda$ could be placed in reproducible fashion as in the equipment of Ref. 2.

The ionization currents, assuming chambers of equal and constant sensitivity, are

$$I_1(t)=I_1(0)e^{-\lambda t}, \quad (1)$$

$$I_2(t)=I_2(0)e^{-(\lambda+\Delta\lambda)t}, \quad (2)$$

provided that the sources are free from any other radioactivity and that no change in position of the source material nor any migration or removal of source material takes place. The difference current is

$$\Delta i(t)=e^{-\lambda t}[\Delta i(0)+I_2(0)\Delta\lambda t], \quad (3)$$

provided that $\Delta\lambda t \ll 1$. In practice, the difference current is obtained directly by collecting positive ions in one chamber and negative ions in the other.[2,3] By interchanging the sources to minimize differences in the sensitivities of the chambers, an average value of the difference currents is obtained referred to time t.[2,9]

For convenience in analysis the difference current is multiplied by $e^{\lambda t}$, yielding

$$e^{\lambda t}\Delta i(t)=\Delta i(0)+I_2(0)\Delta\lambda t, \quad (4a)$$

i.e.,

$$y=a+bt. \quad (4b)$$

In practice, two sets of measurements are made: $I(t)$ versus t, and $\Delta i(t)$ versus t. By least-squares analysis, the values of λ and $I(0)$ are obtained from the first set. The desired result, $\Delta\lambda/\lambda$, is equal to $b/I_0\lambda$.

Depending on their magnitudes, the ionization currents are measured either by the deflection method or by the rate-of-drift method, utilizing a null technique in both cases.[2]

III. SOURCES

A. Preparation

Sources were sought in which the tellurium would be in different valence states, which would be stable against physical or chemical changes, and which could be checked by x-ray analysis. The following sources were selected for use: Te(precipitated element), Ag_2Te (synthetic hessite), and TeO_2.

Te^{125m} in a weak HNO_3 solution was supplied by the Oak Ridge National Laboratory. The material was produced at Oak Ridge by the irradiation of 10 g of natural tin metal. The target was in the reaction continuously for 1268 days. The total integrated irradiation was 2.24×10^{21} neutrons/cm^2. Five months after removal from the reactor the irradiated tin underwent separation and purification as follows: The tin target was dissolved in HCl, and sufficient inert Te carrier was added to give high recovery of the radioactive tellurium isotopes present; the tellurium was separated by precipitation with SO_2, leaving in solution the tin and antimony which had been formed during irradiation; the tellurium was redissolved in HNO_3 and shipped to supply the material used in these experiments.

This material, when analyzed before shipment by ORNL,[10] was found to contain, in addition to the Te^{125m} radiations, only a weak intensity gamma radiation of 160-keV energy associated probably but not positively with Te^{123m}. At the beginning of the experimental work on $\Delta\lambda$ reported here, the activity of the contaminant Te^{123m} was estimated to be roughly 0.04% of the main Te^{125m} source activity. Confirmatory experiments on the identity of the contaminant and its activity will be reported in the next section.

The TeO_2 and Ag_2Te sources were prepared from precipitated Te which contained a suitable activity of Te^{125m} (plus inactive carrier) to the amount of a few mg/mCi.

The precipitated Te was obtained for the above sources and for the Te source by the following procedure. Inactive pure TeO_2 was dissolved in HNO_3. Te^{125m} in weak HNO_3 was added from the ORNL material to yield the proper activity, and the solution was evaporated to dryness. A few milliliters of con-

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[8] E. Rutherford, Wien. Ber. **120**, 303 (1911).

[9] A. C. Malliaris, thesis, Harvard University, 1961 (unpublished).

[10] Private communication from ORNL; courtesy of P. S. Baker and H. Blauer.

centrated HCl were added to the dry salt until the TeO_3 was completely dissolved. It is known that tellurium can be precipitated in elemental form by SO_2 from an HCl solution, provided the acidity is below 6*M*. The solution was therefore adjusted to 3*M* and heated. Just below the boiling point, SO_2 was passed through the solution until the Te was well coagulated. The precipitate was then collected by centrifuging, washed repeatedly, and dried in a vacuum.

The TeO_2 sources were prepared from the Te^{125m} plus carrier by dissolving in HNO_3, heating until all the Te was in solution, evaporating to dryness, and heating gently to expel traces of HNO_3. Then the salt was decomposed to TeO_2 by the addition of boiling water, and the TeO_2 was dried. The process was repeated and then the dry TeO_2 was collected.

The Ag_2Te (synthetic hessite) sources were prepared according to a method of N. L. Markham.[11] Carefully weighed amounts in atomic proportions to form Ag_2Te of the precipitated Te and of pure silver metal as a fine powder were ground together thoroughly. The mix was transferred to a small porcelain boat and sealed in an evacuated Pyrex tube which was heated at 300°C for 4 days. The final intensity was in the range of 300 to 400 μCi per source radium gamma-ray equivalent.

The radioactive sources used in this work were not checked directly by x-ray structure analysis, since their amount and form were not convenient for direct identification and the losses involved would have been intolerable. Instead samples of Te, TeO_2, and Ag_2Te were prepared from the inactive tellurium carrier material by the identical sequence of operations which had been used for the radioactive sources. These inert compounds were analyzed by the x-ray diffractometer. The diffractometer records were read directly and interpreted with the help of crystallographic data, and were also compared to records made from samples of known identity. Te was crystallized in the hexagonal system with $a=4.456$ Å, $c=5.926$ Å; TeO_2 was orthorhombic with $a=5.50$ Å, $b=11.75$ Å, $c=5.59$ Å.[12] The Ag_2Te (synthetic hessite, low-temperature polymorph) was orthorhombic with $a=16.27$ Å, $b=26.58$ Å, $c=7.55$ Å. The cell content equalled 48 Ag_2Te.[11]

Proper protection of the sources must be provided against mechanical and chemical changes for the duration of an experiment which may run for several months. The results of an early set of experiments had to be rejected because a coating of polystyrene used to immobilize the sources, although it was considered satisfactorily resistant to radiation effects, did apparently enter into chemical combination with the Te of the compounds to form small amounts of volatile hydrides which escaped.[9] In the second set of experiments, described here, an aluminum metal case was used to cover and support the sources. The sources were placed in a cup 0.250 in. in diameter with a wall height of 0.050 in., made of Al foil 0.003 in. thick. The source material was placed in the cup and covered with a lid of slightly smaller diameter than the inside of the cup. The wall of the cup was turned down over the lid, and the assembly was pressed so that a thin disk resulted in which the source material was immobilized. This disk was then placed within the flattened end of a thin-walled aluminum tube which was squeezed to hold the disk firmly. The other end of the tube was attached to the source holder which fitted precisely into the re-entrant Dural tubes of the ionization chambers.

The activities of the individual source materials used were roughly matched to 10% before being encapsulated. The absorption of the aluminum tube holders was varied for individual sources to achieve equality of the ionization currents to about one part in a thousand.

B. Radioactive Contaminants

The source material, Te^{125m} in solution in weak HNO_3, was analyzed before shipment by ORNL and was found to contain a weak source of $\approx$160-keV gamma radiation present to about 0.04% of the activity of the Te^{125m}. The contaminant was tentatively identified as Te^{123m}. Following receipt of the source, the material was examined using a NaI(Tl) scintillation spectrometer to identify the contaminant and obtain an independent measurement of its amount.

In agreement with the ORNL results, no gamma rays were found other than the radiation from Te^{125m} and a radiation of 158.4 ± 1.3 keV whose value agrees with that of the 159.1-keV line of Te^{123m}.[13] The period of half-life of the contaminant radiation was determined from measurement of intensity ratios, made at different times during the experiment, relative to Te^{125m}, using the scintillation spectrometer. The half-life of the contaminant was roughly 93 days, in satisfactory agreement with the accepted value of 104 days for Te^{123m}.[13]

Additional evidence for the identification of the contaminant as Te^{123m} was obtained from consideration of the genesis of Te^{125m} and Te^{123m} from a natural tin target containing 6% Sn^{124} and 33% Sn^{120}, irradiated by neutrons. The ratio of activities of Te^{123m}/Te^{125m} may be calculated from the data of the irradiation history of the target as previously summarized and the (n,γ) cross sections and decay characteristics of the radioactive isotopes involved in the genetic sequences. Half-

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[11] N. L. Markham, thesis, Harvard University, 1957 (unpublished).

[12] Tables for conversion of x-ray diffraction angles to interplanar spacings, *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1964), Appl. Math. Ser., Vol. 10; R. W. G. Wyckoff, *Crystal Structures* (Interscience Publishers, Inc., New York, 1960).

[13] E. Der Mateosian and M. McKeown, Brookhaven National Laboratory Report No. BNL 605, 1960 (unpublished).

life periods are used in the following genetic sequences.

$$\underset{0.2\text{ b}}{\mathrm{Sn}^{124}(n,\gamma)\mathrm{Sn}^{125}} \xrightarrow[\text{10 min}]{} \mathrm{Sb}^{125} \xrightarrow[\text{2.7 yr}]{20\%} \mathrm{Te}^{125m} \xrightarrow[\text{58 day}]{} \quad (5)$$

$$\underset{0.14\text{ b}}{\mathrm{Sn}^{120}\ (n,\gamma)\ \mathrm{Sn}^{121}} \xrightarrow[\text{27 h}]{} \underset{7\text{ b}}{\mathrm{Sb}^{121}(n,\gamma)\mathrm{Sb}^{122}} \xrightarrow[\text{2.8 day}]{}$$

$$\underset{1.1\text{ b}}{\mathrm{Te}^{122}(n,\gamma)\mathrm{Te}^{123m}} \xrightarrow[\text{104 day}]{} . \quad (6)$$

The calculated ratio of activities $\mathrm{Te}^{123m}/\mathrm{Te}^{125m}$ for these genetic sequences was $\approx 0.02\%$ referred to the start of the current experiments. This ratio of 0.02%, when compared with the approximate value of 0.04% from ORNL, concluded the series of evidence that the contaminant present in the Te^{125m} source material was indeed Te^{123m}.

One set of measurements of the activity ratio was made by comparing the 160-keV radiation of Te^{123m} to the 110-keV radiation of Te^{125m} originating in the same source. The ratio of activities $\mathrm{Te}^{123m}/\mathrm{Te}^{125m} \approx 0.07\%$, when allowance is made for the fact that only 0.34% of the Te^{125m} activity is associated with the unconverted 110-keV gamma radiation.

In another set of measurements in which the Te^{123m} 160-keV gamma ray was measured from several sources of known Te^{125m} activity, the activity ratio was 0.06%. The four figures 0.02, 0.04, 0.07, and 0.06% were averaged to 0.05% to give a first rough estimate of the systematic error due to the contaminant. On this basis there was 0.18 μCi of Te^{123m} in the average source of 350 μCi of Te^{125}.

More reliable correction for the effect of the contaminant requires that the amounts present in each individual source be known. In this work seven sources were used: $(\mathrm{Ag_2Te})_{\mathrm{I}}$, $(\mathrm{Te})_{\mathrm{I}}$, $(\mathrm{Te})_{\mathrm{II}}$, $(\mathrm{TeO_2})_{\mathrm{I}}$, $(\mathrm{TeO_2})_{\mathrm{II}}$, $(\mathrm{Te})_{\mathrm{III}}$, $(\mathrm{Ag_2Te})_{\mathrm{II}}$. The first five sources were used for the main experiment, with the source materials enclosed in Al cups as described earlier. The last two sources, used in auxiliary measurements, were in 0.004-in. thick pure tin capsules similar in design to the Al cups.

The relative amounts of the contaminant in individual sources were measured by the scintillation spectrometer and are listed in Table I. Chiefly because of the absorption difference between 0.004-in. Sn and 0.003-in. Al for the 27-keV radiation of Te^{125m}, the amount of source material in the tin-covered sources, $(\mathrm{Ag_2Te})_{\mathrm{II}}$ and $(\mathrm{Te})_{\mathrm{III}}$, had to be three or four times greater than in the others, with a corresponding increase in the amount of Te^{123m}. The data of Table I were used to calculate the fraction of each $\Delta i(t)$ contributed by the unbalanced contaminant. The conversion coefficient relating scintillation data to chamber ionization currents was determined as described in the next section.

TABLE I. Scintillation spectrometer measurements of the relative amount of the contaminant activity (arbitrary units) in individual sources at $t=0$. The conversion factor $=0.0154\pm0.0010$ to convert to chamber ionization current. The figures listed have a standard deviation $\approx 3\%$.

Source No.	Composition and identification number	Activity
1	$(\mathrm{Ag_2Te})_{\mathrm{I}}$	667
2	$(\mathrm{Te})_{\mathrm{I}}$	605
3	$(\mathrm{Te})_{\mathrm{II}}$	662
4	$(\mathrm{TeO_2})_{\mathrm{I}}$	663
5	$(\mathrm{TeO_2})_{\mathrm{II}}$	622
6	$(\mathrm{Te})_{\mathrm{III}}$	2119
7	$(\mathrm{Ag_2Te})_{\mathrm{II}}$	2630

IV. CORRECTION FOR THE EFFECT OF THE CONTAMINANT

The contaminant, as an isotope of the main source, could not be eliminated by chemical separation. Its effect was taken into account by subtracting its contribution to the ionization currents as detailed below.

The presence of a contaminant with decay constant λ' in the sources 1 and 2 will add terms $J_1(0)e^{-\lambda' t}$ to Eq. (1), and $J_2(0)e^{-\lambda' t}$ to Eq. (2). Then, following the procedure which led to Eqs. (2), (3), (4), and (4a),

$$e^{\lambda t}\Delta i' = \Delta i(0) + I_0 \Delta\lambda t + \Delta j(0)e^{(\lambda-\lambda')t}, \quad (7)$$

and

$$y' = e^{\lambda t}\Delta i' = a + bt + \Delta j(0)e^{(\lambda-\lambda')t}. \quad (8)$$

The values of $J(0)$ and $\Delta j(0)$ for the individual sources were obtained by measuring the time dependence of the difference currents for the three pairs of sources $(\mathrm{Ag_2Te})_{\mathrm{II}}$–$(\mathrm{Ag_2Te})_{\mathrm{I}}$, $(\mathrm{Te})_{\mathrm{III}}$–$(\mathrm{Te})_{\mathrm{I}}$, and $(\mathrm{Te})_{\mathrm{III}}$–$(\mathrm{Te})_{\mathrm{II}}$. In the case of paired sources of identical chemical composition, since $b=0$, the results of the measurements can be represented from Eq. (8) as

$$y' = a + \Delta j(0)e^{(\lambda-\lambda')t}.$$

As λ and λ' are known, it is possible to solve for $\Delta j(0)$. The three ionization measurements of $\Delta j(0)$ from the three pairs of sources were then used to obtain the conversion coefficient which related the scintillation spectrometer activities listed in Table I to the ionization chamber values of $J(0)$ and $\Delta j(0)$. The average value of the conversion coefficient was 0.0154 ± 0.0010 ionization current divisions (10^{-14} A) per unit of contaminant activity listed in Table I. This value supplanted an earlier calculated value of 0.013 for the conversion coefficient. The differences of contaminant activities $\Delta j(0)$ are listed in the third column of Table II, which also gives the results before and after correction for the effects of the contaminant. The small effect of the contaminant on the direct determination of λ will be discussed under Sec. V.

The general case of radioactive contaminants in experiments of this kind is considered in detail in Ref. 9.

V. RESULTS

The decay constant λ of Te^{125m} and the initial current values $I(0)$ were obtained by measuring the decay of

TABLE II. The difference ionization currents $\Delta j(0)$ (due to the contaminant), and the quantities a and b before and after correction for the systematic error due to the contaminant.

				Uncorrected for contaminant				Corrected for contaminant			
Ref. No.	Source pair	$\Delta j(0)\pm\sigma(\Delta j)$ divisions		$a\pm\sigma(a)$ divisions		$b\pm\sigma(b)$ 10^{-3} div/day		$a\pm\sigma(a)$ divisions		$b\pm\sigma(b)$ 10^{-3} div/day	
1	$(Te)_{II}-(Te)_I$	0.88	0.42	5.87	0.16	5.91	3.60	5.09	0.45	0.05	4.17
2	$(TeO_2)_{II}-(TeO_2)_I$	−0.63	0.42	1.98	0.18	−4.03	3.94	2.59	0.46	0.31	4.46
3	$(Ag_2Te)_I-(Te)_I$	0.95	0.42	8.80	0.19	39.99	4.40	7.87	0.46	33.72	4.88
4	$(Ag_2Te)_I-(Te)_{II}$	0.0	0.43	2.51	0.13	35.39	3.03	2.51	0.45	35.39	3.71
5	$(Ag_2Te)_I-(TeO_2)_I$	0.0	0.43	10.40	0.16	27.92	3.68	10.40	0.46	27.92	4.26
6	$(Ag_2Te)_I-(TeO_2)_{II}$	0.69	0.42	8.09	0.16	37.09	3.68	7.41	0.45	32.58	4.24
7	$(Te)_I-(TeO_2)_I$	−0.89	0.42	2.08	0.17	−13.29	4.89	2.97	0.45	−7.91	5.32
8	$(TeO_2)_{II}-(Te)_I$	0.26	0.40	0.35	0.21	7.89	5.37	0.09	0.45	6.28	5.73
9	$(Te)_{II}-(TeO_2)_I$	0.0	0.43	7.82	0.18	−3.27	4.45	7.82	0.47	−3.27	4.94
10	$(Te)_{II}-(TeO_2)_{II}$	0.62	0.43	5.60	0.21	0.70	5.14	4.99	0.48	−3.29	5.57

source No. 2 $(Te)_I$ and source No. 5 $(TeO_2)_{II}$. The least-squares results, corrected for the presence of the contaminant, are for source No. 2: $I(0)=11\,233$ divisions $\pm 0.06\%$, and $\lambda=0.01201$ day^{-1} $\pm 0.07\%$; and for source No. 5: $I(0)=11\,234$ divisions $\pm 0.05\%$, and $\lambda=0.01201$ day^{-1} $\pm 0.07\%$. Errors are given as standard deviations throughout.

The contribution of the contaminant $J(0)\approx 10$ divisions may be compared to the nominal average main source strength $I(0)=11\,240$ divisions (10^{-14} A). If uncorrected for the presence of the contaminant, the error would be approximately $\lambda/2000$ in determining λ from $I'(t)=I(0)e^{-\lambda t}+J(0)e^{-\lambda' t}$ observed over two periods of half-life of the main source.

Table II summarizes the results of the intercomparison of sources, listing the initial difference in intensity $\Delta i(0)=a$ and the slope $I(0)\Delta\lambda=b$ associated with Eqs. (4) and (4a). $\lambda=0.0120$ day^{-1} was used in calculating the exponential factor in the term $e^{\lambda t}\Delta i(t)$. Each individual measurement was corrected for the presence of the contaminant and a new least-squares solution was obtained. Therefore the corrected values of a are not simply the uncorrected values of $a-\Delta j(0)$.

Figures 2, 3, and 4 are plots of $y=e^{\lambda t}\Delta i(t)=a+bt$ for six of the pairs of sources listed in Table II. The solid lines represent the least-squares solution of the data for which the values of a and b are given in the last two column entries of Table II. Figure 2 illustrates one method of checking the over-all performance of the apparatus, as within the errors essentially zero slope was recorded for pairs of sources of identical chemical composition.[2]

Other checks on the measurements are provided by a comparison of a and b values for ten sets of three pair measurements from Table II. For any three individual sources generalized as A, B, and C, the respective values

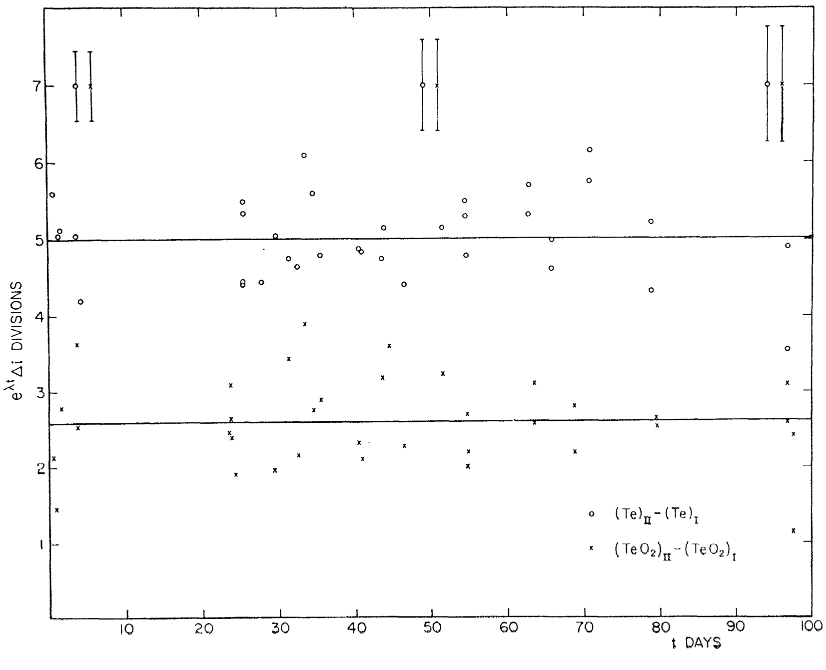

FIG. 2. Data on pairs of sources of identical composition, pair 1 (Te) and pair 2 (TeO_2) of Table II, which were measured as a test of the over-all operation of the apparatus. The solid lines represent the least-squares solutions of the data. The ordinates are in units of 10^{-14} A = 1 division. The intensity of the sources was $I(0)\approx 11\,240$ divisions at $t=0$. Sample standard deviations are indicated at the top of the plot.

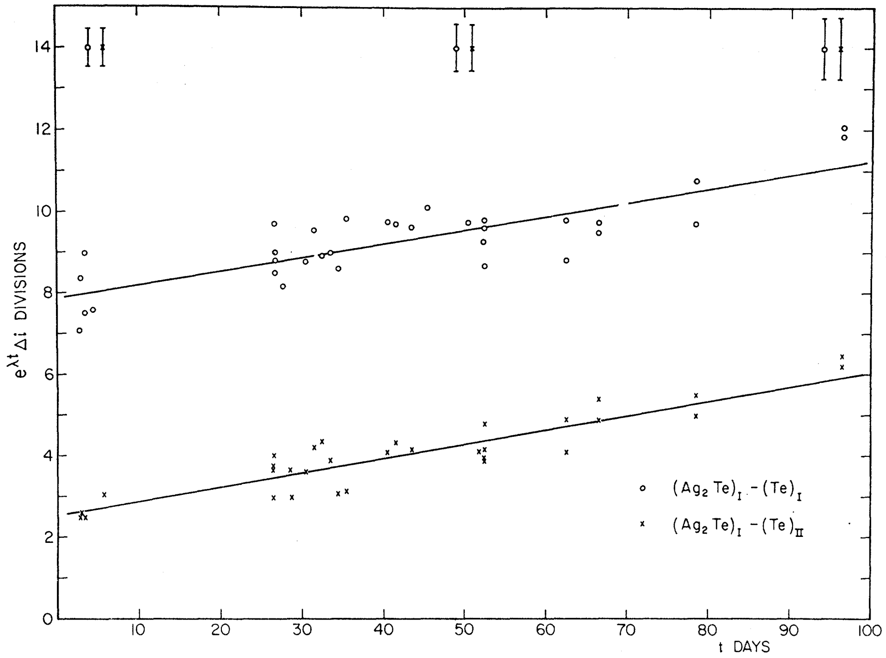

FIG. 3. Pairs 3 and 4 of Table II illustrate the greater decay rate of Te with respect to Ag_2Te.

of *a* and *b* should agree within the distribution of errors for $(A-C)$ compared to $(A-B)-(C-B)$. These comparisons were satisfactory.

The calculation of the final values for $b=I(0)\Delta\lambda$, from which $\Delta\lambda/\lambda$ was obtained, was made as follows: Properly weighted averages were found to yield average values of *b* from pairs 3 and 4, (Ag_2Te)–(Te); from 5 and 6, (Ag_2Te)–(TeO_2); and from 7, 8, 9, and 10, (Te)–(TeO_2). These values of *b* were, respectively, 34.78±2.95, 30.26±3.00, and $-5.11\pm2.68\times10^{-3}$ div/day. Then a constraint fitting was applied, using a least-squares procedure, following the method of Deming.[14] The constraint was that the sum of the first and third values of *b* must equal the second value of *b*.

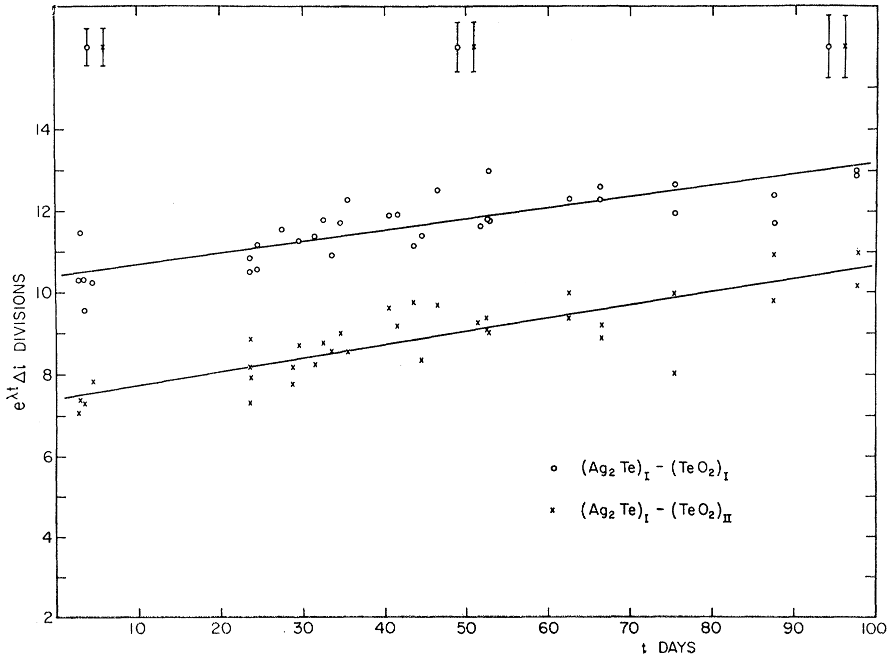

FIG. 4. Results on source pairs 5 and 6 of Table II.

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[14] W. E. Deming, *Statistical Adjustment of Data* (John Wiley & Sons, Inc., New York, 1946).

The results of the constraint fitting are: $34.99 \pm 2.38 \times 10^{-3}$ div/day for (Ag_2Te)–(Te); $30.05 \pm 2.40 \times 10^{-3}$ div/day for (Ag_2Te)–(TeO_2); and $-4.94 \pm 2.26 \times 10^{-3}$ div/day for (Te)–(TeO_2).

These final slope values determined the fractional change in λ from the relation $\Delta\lambda/\lambda = b/I(0)\lambda$, where $I(0) = 11\,240$ divisions and $\lambda = 0.0120$ day^{-1}. The values for $\Delta\lambda$ are: $\lambda(\mathrm{Te}) - \lambda(\mathrm{Ag_2Te}) = 2.59 \pm 0.18 \times 10^{-4} \lambda(\mathrm{Te})$; $\lambda(\mathrm{TeO_2}) - \lambda(\mathrm{Ag_2Te}) = 2.23 \pm 0.18 \times 10^{-4} \lambda(\mathrm{Te})$; and $\lambda(\mathrm{Te}) - \lambda(\mathrm{TeO_2}) = 0.36 \pm 0.17 \times 10^{-4} \lambda(\mathrm{Te})$.

The small effect of different chemical environments on the transition rate of Te^{125m} as compared with the results obtained from similar experiments on Tc^{99m} is not unexpected. In the present case, a 109-keV transition is involved, and the electrons more amenable to chemical influence contribute less to the total transition rate than in the case of Tc^{99m}, where only a 2-keV transition was involved.[2]

The smaller standard deviations associated with the present results for Te^{125m} derive from two factors. First, many more measurements were possible for a substance of 58 days half-life than for Tc^{99m}, which has a half-life of 6 h. Second, the number of ionization events contributing to approximately the same values of $I(0)$ was somewhat greater for Te^{125m} with a 27-keV radiation than for Tc^{99m}, with a radiation of 140 keV.

The number of ionization events per second N which contributed to the initial current $I(0) = 11\,240 \times 10^{-14}$ A was approximately 720 000. This calculation was based an absorption of 25 keV per ionization event and on 25 to 28 eV per ion pair produced. The duration of a single measurement of a different current was 1000 sec, and in the initial measurements approximately 7.2×10^8events occurred in each chamber. $N(t) = N(0)e^{-\lambda t}$ at time t. The relative standard deviation of the difference currents $\Delta I(t)$ was $\sigma[\Delta i(t)]/I(t) = N(0)^{-1/2} e^{\lambda t/2}$ for the average of two measurements with the sources interchanged in the chambers.

More than 85% of the magnitude of the final error in $\Delta\lambda/\lambda$ arises from this statistical fluctuation in the number of ionizing events. The other 15% comes mainly from the uncertainty $\sigma[\Delta j(0)]$ in the correction for the contaminant Te^{123m}. The standard deviation in the slope associated with error in $\Delta j(0)$ equals $\sigma[\Delta j(0)] \times (\lambda - \lambda') e^{(\lambda - \lambda')t}$ by differentiation of Eq. (8). This deviation of 2×10^{-3} div/day initially at small values of t decreases to $\sim 1.2 \times 10^{-3}$ div/day at the end of the experiment, $t \approx 100$ days, and contributes less than 15% to the final error in $\Delta\lambda/\lambda$.

The following possible sources of error were also considered, but their effects on $\Delta\lambda/\lambda$ are negligible: $\sigma(\lambda) \leqslant 0.1\%$; lack of ion current saturation $< 1\%$; leak rate of argon in the chambers $< 0.5\%$/yr; background ionization current per chamber $< \pm 1$ division; differential background current < 0.05 divisions. The background currents listed include the effects of local radioactivity and cosmic radiation, electrometer drift, and currents induced by variations in the ion collecting potentials.

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