Angular Correlation of Gamma Rays in Pb^{204m}†

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The anisotropies of the three pairs of gamma rays in the triple cascade of 1.1-hour Pb^{204m} have been measured. The results were 0.163±0.010 for the 374–899 kev (prompt) pair; 0.414±0.015 for the 913–899 kev pair; and 0.398±0.011 for the 913–374 kev pair. These results are consistent with the scheme, 9(*E*5)4(*E*2)2(*E*2)0.

INTRODUCTION

THE multipole character of the gamma rays of the 1.1-hour Pb^{204m} cascade (Fig. 1) has been in question for some time. In order to reconcile the angular correlation measurements[1] of the delayed (913–374 kev and 913–899 kev) gamma-ray pairs with the existing internal conversion data[2] it was necessary to assume that the 374-kev gamma ray was a mixture of *E*2 with 0.5% *M*3 and that the 913-kev radiation was a mixture of *E*5 with 1% *M*6. As it is difficult to understand such mixtures on the basis of any of the fashionable nuclear models, it seemed desirable to perform additional experiments to check the consequences of the above-mentioned assumptions. The assumed mixtures[1] led to a prediction of 0.23 for the anisotropy of the prompt (374–899 kev) gamma-ray pair whereas a pure 4(*E*2)2(*E*2)0 transition would have an anisotropy of 0.167. Therefore, a study of the anisotropy of this pair was undertaken. The results of this study indicated that a repetition of the measurements on the delayed (913–374 kev and 913–899 kev) pairs, with sources prepared in a manner different from the previous method[1] was in order.

374-899 kev Cascade

The Pb^{204m} sources made by deuteron bombardment of thallium foils[1] could not be used for the prompt-pair measurement because other lead activities, which had coincidences of gamma rays of similar energy, were produced; so two other methods were used to prepare sources.

First, lead oxide, enriched in Pb^{204} to about 27%, was placed in the fast-irradiation facility of the Argonne Research Reactor, CP-5. The 2.2-Mev isomeric state in Pb^{204} was excited by the inelastic scattering of fast neutrons. The anisotropy of the 374–899 kev pair was measured with the electronic apparatus previously described.[1] The fast-coincidence circuit had a resolution time of the order of 5×10^{-8} sec. The source was not dissolved in any liquid, for the intermediate half-life is less than 6×10^{-10} sec [3] which should be sufficiently short to preclude any detectable attenuation due to interactions with the surrounding medium.[4]

The 899-kev gamma ray was detected by a NaI(Tl) crystal 2.5 inches in diameter and 2 inches thick with a 0.125-inch lead absorber in front of it. The counter circuit was biased to select pulses caused by radiation above 600 kev. The 374-kev gamma ray was detected by a NaI(Tl) crystal, 1.5 inches in diameter and one inch thick. A tantalum absorber 0.015 inch thick was used on the face of this crystal. The differential pulse-height analyzer in this channel was set with a window which detected pulses corresponding to radiation of 300 to 450 kev. The decay of the coincidences obtained with this scheme and the absence of strange peaks in the pulse-height distribution indicated that undesired activities were not present in the sources.

The data had to be corrected for coincidences caused by the 913–374 kev and the 913–899 kev pairs. The 913–374 kev gamma-ray pair have an intermediate half-life of 2.6×10^{-7} sec and make a 13% contribution to the total number of coincidences. The anisotropy of this pair in the solid PbO source was measured by delaying the appropriate counter by 1.9×10^{-7} sec so that prompt coincidences were not observed. Some pulses caused by the 898-kev gamma ray appear in the 300- to 450-kev window. This contribution was measured by delaying the other counter 8.1×10^{-8} sec and using the fact that the first and third gamma rays have almost the same energy and the same intensity. The measurement indicates that the ratio of coincidence from the 912–899 kev cascade to the 913–374 coincidence was 12% for our bias conditions. Under the delay conditions of our measurement, the measured anisotropies of the 913–374 and 912–899 cascades were combined with the relatively small percentages of each present, 13 and 2%, respectively, to give a correction

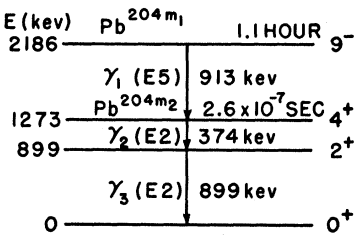

FIG. 1. Pb^{204m} decay scheme.

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of +0.010 to the measured anisotropy of the 374–899 kev cascade. An anisotropy of 0.156±0.014 was obtained as a result.

The second measurement of the anisotropy of the 913–374 kev gamma-ray pair was made with carrier-free sources of Pb^{204m}. Ordinary lead was melted into a depression, 0.013 cm deep, 1 cm wide, and 10 cm long, in a copper cyclotron target equipped for water cooling. The individual lead targets were bombarded in the Argonne Cyclotron with approximately 1000 μa-hours of 22-Mev deuterons producing several bismuth isotopes. A few hours after the end of the bombardment the lead was melted out of the groove in the copper target and dissolved in hot 5N HNO_3. Concentrated HCl was added to precipitate most of the lead. The supernatent was evaporated to almost dryness and made 1N in HCl (additional $PbCl_2$ precipitate was separated by centrifugation). The above 1N HCl solution was stirred with nickel powder (approximately 500 mg) for about one-half hour at 80 to 90°C in order to absorb the carrier-free bismuth on the nickel.[5] The nickel was centrifuged off, washed, and dissolved in hot 6N HNO_3. One mg of ferric iron was added to the latter solution and the iron was precipitated with excess ammonia to carry the bismuth activity. The ferric hydroxide was dissolved and the precipitation was repeated twice with washes to remove the nickel. The final ferric hydroxide precipitate (containing the bismuth) was dissolved in 6N HCl and the iron removed by ethyl ether extractions. The remaining solution was diluted to about 1N HCl and placed on a Dowex Al anion resin column. The 1.1-hour Pb^{204m}, daughter of 12-hr Bi^{204}, was eluted off the resin column with 0.1N HCl, the bismuth being strongly absorbed on the anion resin at this acid concentration. The first source of Pb^{204m} was available approximately 12 hours after the end of the cyclotron bombardment.

Pb^{203} is also produced by the reaction $Pb^{204}(d,3n)Bi^{203}$ followed by the subsequent decay of Bi^{203} to Pb^{203} by electron capture with a half-life of 12 hours. The Pb^{203} activity is small compared to the Pb^{204m} activity produced by the reactions $Pb^{206}(d,4n)Bi^{204}$ and $Pb^{204}(d,2n)Bi^{204}$ with the subsequent decay (12-hour half-life) to Pb^{204m}, because the abundance of Pb^{206} is several times the abundance of Pb^{204} in ordinary lead and the the $(d,4n)$ cross section compares to the $(d,3n)$ cross section for 22-Mev deuterons. The Pb^{204m}/Pb^{203} activity ratio was further favored by eluting the lead from the column containing the bismuth parent about every two hours so that the Pb^{203} was allowed to grow to only a small fraction of its equilibrium decay rate.

A pulse-height analysis of the radiations in the sources, a measurement of the intermediate half-life of 2.6×10^{-7} sec, the observed decay (1.1 hour) of the coincidences accepted by the analyzers, and the favorable ratio of true to accidental coincidences, indicated that there were no interfering activities in the sources.

The liquid sources were boiled down and the final concentration of the HCl solutions was about 6N. The anisotropy of the 374–899 kev cascade was measured in a manner similar to that described above, except that a NaI(Tl) crystal 3.5 inches in diameter and 3.5 inches thick was used to detect the 899-kev gamma-ray and a NaI(Tl) crystal 2.5 inches in diameter by 2 inches thick was used for the 374-kev gamma ray. The correction for the contributions of the delayed pairs (−0.019) was obtained by measuring the anisotropy of the 913–899 kev pair and the 913–374 kev pair in the same liquid sources. The result obtained from this measurement of the anisotropy of the prompt pair was 0.170±0.014. This result was averaged with that obtained from the sources made by inelastic scattering of neutrons and 0.163±0.010 was obtained as our best value for the anisotropy of the 374–899 kev pair. This result is consistent with the decay scheme $4(E2)2(E2)0$.

DELAYED COINCIDENCES

Since this result differs from the prediction based on the results of reference 1, the measurements of the anisotropies of the 913–374 kev pair and the 913–899 kev pair were repeated. The sources were prepared by bombarding ordinary lead with 22-Mev deuterons and following the chemical procedures outlined above. The delayed cascades were studied with one 3.5-inch and one 2.5-inch crystal. The coincidence resolution time was increased to 2.3×10^{-7} sec. For the 913–899 kev cascade a lead absorber 0.125 inch thick was placed on the front of each crystal and the first counter was delayed by about 3.1×10^{-7} sec. A window from 730 kev to 1070 kev was used with each counter circuit. The result for the anisotropy of this pair of gamma rays was 0.414±0.015.

For the 913–374 kev gamma ray pair, the bias conditions on the second counter were changed so that the window of the differential pulse-height analyzer extended from 310 to 430 kev and the lead absorber in front of the second crystal was replaced by 0.015 inch of tantalum. This measurement had to be corrected for the contribution from the 913–899 kev pair, since Compton processes of the 899-kev gamma rays appear in the 310–430 kev window and produce true delayed coincidences. The contribution of this pair (10%) was measured by delaying the second counter by 3.1×10^{-7} sec. The anisotropy of the 913–899 kev pair is not the proper number to use in this correction, however, for true 913–899 kev delayed coincidences can be produced if the 899-kev gamma ray is scattered into the second counter and then produces a pulse which is within the limits of the analyzer window. An auxiliary experiment to determine this correction was performed using Co^{60} as a source. It showed that the anisotropy of the 913–899 kev pair would be reduced 30% when observed with the windows used for the 913–374 kev measurement. The

[5] D. E. Alburger and G. Friedlander, Phys. Rev. **81**, 523 (1951).

correction to the measured 913–374 kev pair anisotropy was +0.011 and the final corrected result was 0.398±0.011. This is, within the experimental error, the same as the anisotropy for the 913–899 kev pair, and both numbers are consistent with 0.408 which is the calculated anisotropy of the first and second and the first and third gamma rays of the scheme 9(*E*5)4(*E*2)2(*E*2)0.

The previous result[1] for the anisotropy of the 913–374 kev pair was 0.34±0.02 which is inconsistent with the above scheme. Efforts to find fault with this measurement have been unsuccessful. Revision of the correction for the contribution of the 913–899 kev pair to conform to the method used in the present experiment raises this result to 0.35±0.02 which is a trivial change. However, the present result is more precise and is consistent with the 9(*E*5)4(*E*2)2(*E*2)0 scheme which explains the other anisotropy measurements and the internal conversion coefficients.[2] Hence, we feel justified in ignoring the previous measurement.

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Atomic Masses from C^{12} to Ne^{22}†

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Doublets leading to mass values of the stable isotopes from C^{12} to Ne^{22} have been measured with a double-focusing mass spectrometer. In the determination of C^{12}, two new cycles have been used, $(\frac{1}{2}C_4H_4O-O^{18}O)-(H_3O-HO^{18})$ and $(\frac{1}{2}C_4H_2O-O^{17}O)-(H_3O-H_2O^{17})$, resulting in the value $C^{12}=12.003814\pm6$ amu.

An analysis of the available data pertinent to the investigated mass region is presented in an effort to uncover specific measurements most likely responsible for the discrepancies in mass values reported from the various sources of investigation. It is found that a generally improved agreement is obtained if adjustments are made in the following reactions: $O^{18}(p,\alpha)N^{15}$, $F^{19}(d,\alpha)O^{17}$, $Ne^{21}(d,\alpha)F^{19}$, and $Ne^{20}(d,\alpha)F^{18}$.

INTRODUCTION

THIS paper presents the results of mass measurements of the stable isotopes from $A=12$ to 22 undertaken with a double-focusing mass spectrometer developed at the University of Minnesota. Among the light nuclides, the list of precise nuclear reaction energy measurements is extensive.[1] Upon such data, generally overdetermined, two different computations, the first by Li *et al.*[2] and Li,[3] the other by Wapstra,[4] have given closely similar results. Substantially less abundant are the data in the present mass region reported by mass spectroscopic investigators[5] who, except for Ewald[6] and Ogata and Matsuda,[7] have confined their measurements in this region to C^{12} and N^{14}. The randomness in the pattern of agreement or lack of it found on comparing Ewald's and Ogata's results with each other as well as with nuclear values (see Tables I and II) leaves inconclusiveness, to an extent greater than desirable, at most of these mass values. More disturbing has been the divergence in values reported for the mass spectroscopic secondary standard C^{12}, where the nuclear value is lower than any mass spectroscopic value.[5]

The need for more mass spectroscopic data is clearly indicated; in particular, since the errors associated with nuclear mass determinations are cumulative, it would appear that a critical comparison with nuclear data ought to be most sensitive in the vicinity of the O^{16} standard. Such a comparison will be presented in this report in an effort to indicate specific measurements considered most likely responsible for the major existing discrepancies.

THE INSTRUMENT

A schematic drawing of the present instrument is given in Fig. 1. The second of its kind constructed in this laboratory, it is essentially a duplicate of the mass-measuring spectrometer described in previous reports.[8–10] The only alteration in instrument design has

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