

Precision Determination of the Energy of the Gamma Ray of Potassium-40

BEROL L. ROBINSON

Western Reserve University,* Cleveland, Ohio
and

Soreq Research Establishment, Israel Atomic Energy Commission, Rehovoth, Israel†

(Received 24 December 1963)

The gamma ray of potassium-40 was compared with the 1368-keV line of sodium-24 in a scintillation spectrometer. The lines of yttrium-88 were simultaneously recorded, and linear interpolation between these lines was corrected to bring the sodium-24 value into accord with the accepted standard value. A proportional correction was applied to the interpolated value for the energy of the potassium-40 line. Sources consisted of (i) yttrium-88 mixed with potassium bicarbonate, and (ii) yttrium-88 mixed with sodium bicarbonate, with the sodium activated by a small neutron source. About seven kilograms of bicarbonate were used in each case; to obtain sufficient potassium-40 activity in (i), and to simulate approximately the small-angle Compton scattering in a substance of equal density in (ii). Full-energy peaks in the pulse-height spectra were fitted to a normal distribution by an iterative method of least squares. With about 100 000 counts in each peak, energy determinations were reproducible to about 0.5 keV. The energy of the potassium gamma ray is 1459.7 ± 1.0 keV. The dependence of the result upon changes in the standards is also given.

INTRODUCTION

THE decay scheme of potassium-40 was set forth by Morrison,¹ and the only addition to it has been the recent discovery² of a very weak (about 0.001%) positron branching to the ground state of argon-40, and the inference of about 0.16% electron-capture branching to the ground state. Because of their great geochronological importance, much effort has been expended lately upon accurate determinations of the absolute and relative decay rates of the branches, but the other details of the decay scheme have received less attention.

The energy of the beta decay³ to the ground state of calcium-40 has been confirmed by mass spectroscopy,⁴ but discrepancies have been noted between the mass spectroscopic value for the argon-potassium-40 mass difference and values deduced from the observation of the (*p,n*) thresholds in argon-40,⁵ and from the measurement of the specific activity of potassium for the emission of argon-K x rays⁶; the electron-capture energy deduced from the latter⁷ must be added to the energy of the first excited state of argon-40 in order to obtain the mass difference. We have performed a precision determination of the gamma-ray energy to help clarify this matter.

Previous measurements of the gamma-ray energy and of the energy of the first excited state in argon-40 are

tabulated in the *Nuclear Data Sheets*.⁸ The most precise single result is that of Good,⁹ who performed an experiment similar to the one reported here; however, he used a source arrangement which might have given rise to a systematic error comparable in magnitude to the probable error assigned to the final result. The experiment described here is a precision comparison of the gamma ray of potassium-40 with the nearby well-known secondary standard line of sodium-24.

INSTRUMENTS

The instrument used was a simple sodium-iodide scintillation spectrometer. During the development of the experimental techniques, several counters were used; the final data were taken with a Harshaw integral line detector consisting of 3-in×3-in. sodium-iodide crystal and a Dumont 6363 phototube. Various power supplies, amplifiers, and pulse-height analyzers were also used.¹⁰ The final form of the experiment was developed to take into account the well-known nonlinearity of the energy response of sodium iodide in the region above one MeV,¹¹ and it was such that nonlinearities in the amplifier and in the analog-to-digital converter of the multichannel analyzer could be lumped together into an over-all nonlinearity of the entire system. Therefore, no special pains were taken to assure or even to examine carefully the linear response of the electronics, other than to be sure that the signals were well within the capability of

* The work at Western Reserve University was supported in part by the National Science Foundation. A preliminary report was given at the Washington meeting of the American Physical Society, April 1963; *Bull. Am. Phys. Soc.* **8**, 321 (1963).

† On leave of absence from Western Reserve University during the academic year 1961-1962.

¹ P. Morrison, *Phys. Rev.* **82**, 209 (1951).

² D. W. Engelkemeir, K. F. Flynn, and L. E. Glendenin, *Phys. Rev.* **126**, 1818 (1962).

³ L. Feldman and C. S. Wu, *Phys. Rev.* **87**, 1091 (1952).

⁴ C. F. Giese and J. L. Benson, *Phys. Rev.* **110**, 712 (1958).

⁵ R. E. Holland and F. J. Lynch, *Phys. Rev.* **113**, 903 (1959).

⁶ J. Heintze, *Z. Naturforsch.* **9a**, 469 (1954).

⁷ B. L. Robinson and R. W. Fink, *Rev. Mod. Phys.* **32**, 117 (1960).

⁸ *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, D. C.), NRC 59-4-40, -41.

⁹ M. L. Good, *Phys. Rev.* **81**, 891 (1951).

¹⁰ The final data were taken with the following instruments: Model 404M power supply (John Fluke Manufacturing Company, Inc., Seattle, Washington); Models 101 amplifier and 201 pre-amplifier (Cosmic Radiation Labs. Inc., Bellport, Long Island, New York); Model 401 multichannel analyzer (Technical Measurements Corporation, West Haven, Connecticut).

¹¹ D. Engelkemeir, *Rev. Sci. Instr.* **27**, 589 (1956); also C. D. Zerby, A. Meyer, and R. B. Murray, *Nucl. Instr. Methods* **12**, 115 (1961); J. Kantele and R. W. Fink, *ibid.* **13**, 141 (1961).

the main amplifier and the input inverter of the multi-channel analyzer, and that there was no gross non-linearity. The latter was checked with a mercury switch pulser which depended upon a helical potentiometer for its output control.¹² The final experiments were carried out in an air-conditioned laboratory, but no other efforts were made to control the temperature of the system. It is surmised that the residual drifts observed may have been due in part to temperature variations during the course of the experiments.

The source and detector were shielded by at least two inches of lead. Forty-minute counting periods were required to accumulate data of adequate statistical significance.

SOURCES

On account of the extremely low specific activity of potassium for the emission of the argon-40 gamma ray, one can ordinarily obtain adequate counting rates only by surrounding the scintillation detector with a rather massive source, typically five kilograms of potassium. In our earliest experiments we found that the response of the scintillation spectrometer to a point source of monochromatic radiation depended upon the location of the point source with respect to the massive potassium source. In particular, the "peak-to-valley" ratio of the pulse-height spectrum was decreased as the point source was moved from a position adjacent to the detector to a position behind the massive potassium source. When expressed in this way, it seemed quite clear that the response was degraded by small-angle Compton scattering in the potassium source, and that the average pulse height of the full-energy peak might well be shifted to lower values.

An attempt was made to avoid this situation by using a source of potassium highly enriched in potassium-40. The source might then be thinner, and concentrated closer to the detector. Our experience with an enriched source is described in the Appendix.

Since the limited resolving power of the scintillation spectrometer does not permit the simultaneous observation of both the potassium-40 and the sodium-24 lines, we used a method of alternating sources. A small amount of sodium-24 was mixed intimately with a quantity of sodium chloride comparable in mass and arrangement to the potassium source. Potassium hydroxide was chosen for the latter, as having about the same density and scattering characteristics as sodium chloride. With this source arrangement, a number of runs were made alternating potassium-40 and sodium-24 sources. Both lines of the sodium spectrum were recorded and located by a method of least squares as described below, and an attempt was made to interpolate the energy of the potassium-40 line according to the energy scales established by the preceding and following sodium-24 spectra.

Although the experiments were performed in an air-conditioned building, with care taken to maintain the orientation of the phototube with respect to the (magnetic field in the) laboratory, and to adjust the counting rate due to the sodium-24 to be very nearly the same as that due to the potassium-40; uncontrollable and apparently random drifts were observed, and it was quite impossible to obtain results of any value at all.

Although the lines of yttrium-88 at 900 and 1840 keV can be readily resolved from the potassium-40 line at 1460 keV, a linear-energy interpolation in the pulse-height spectrum of a mixture of these activities cannot be made on account of the nonlinear energy response of sodium iodide in this region. A number of attempts have been made to characterize this nonlinearity, but they have been limited in their precision in part due to residual nonlinearities of the amplifiers and pulse-height analyzers used to make the measurements. Even if there were good agreement on the character of the nonlinearity, the use of this information would require equally precise knowledge of the nonlinearity of one's own amplifier and kick sorter. However, by imposing standard reference lines upon both the unknown spectrum and the comparison standard, one can effectively take all the nonlinearities together in one over-all nonlinear calibration curve, provided only that reasonable care be taken to see that the pulse-height spectrum is kept in the same region of the multichannel analyzer and that there are no gross nonlinearities in the system. The issue of long-term pulse-height stability is also evaded.

In final form, the experiment consisted of taking a series of alternate spectra of similarly massive sources of potassium-40 and sodium-24, each containing a uniformly distributed admixture of an appropriate amount of yttrium-88, so that each full-energy peak stood clearly above the Compton distribution of the next higher line. The potassium was in the form of bicarbonate; the sodium source consisted of a similar amount of sodium bicarbonate, which fortuitously has almost the same density, arranged in a similar geometry, as shown in Fig. 1. The sodium was activated by placing a 200 mg radium-beryllium neutron source in the cavity. When the neutron source was surrounded with about 500 g of water, the activation of the sodium was enhanced manyfold,¹³ and its equilibrium activity exceeded the natural activity of potassium.

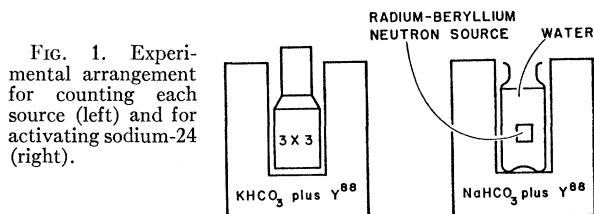


FIG. 1. Experimental arrangement for counting each source (left) and for activating sodium-24 (right).

¹² Model 370 precision pulse generator (ORNL Q-1212 pulser) (Franklin Electronics, Inc., Bridgeport, Pennsylvania).

¹³ E. Amaldi, O. D'Agostino, E. Fermi, B. Pontecorvo, F. Rasetti, and E. Segrè, Proc. Roy. Soc. (London) **A149**, 522 (1935).

TABLE I. Linear interpolations and corrections.

	Sodium-24	Potassium-40
	1378.74 keV	1471.07 keV
	79.64	0.69
	79.73	1.24
	80.29	1.38
	80.83	1.82
Average and probable error	1379.85±0.70	1471.24±0.37
Standard	1368.41±0.20	
Nonlinearity correction	-11.44±0.73	-10.87±0.73
Compton edge correction		1460.37±0.81 -0.5 ±0.2 1459.87±0.84

TREATMENT OF THE DATA

After subtraction of the background and the assumed constant contribution due to the Compton distribution of higher energy gamma rays, the data in each full-energy peak were fitted to a Gaussian distribution by a method of least squares.¹⁴ The statistical error in locating each peak, due only to the finite numbers of events in each channel, was about 0.5 keV. The centroids of the three full-energy distributions of each spectrum were used to make *linear* interpolations of the sodium-24 and potassium-40 gamma-ray energies between the yttrium-88 lines at 897.5 ± 0.5 and 1836.6 ± 0.9 keV.¹⁵ On account of the nonlinearity of the system, the result for sodium-24 obtained in this fashion does not in general agree with the established value of 1368.41 ± 0.20 keV,¹⁶ and a correction must be made, which is in fact a measure of the nonlinearity of the system. A proportional correction was applied to the interpolated result for potassium-40. Assuming a quadratic calibration curve, the correction for the potassium-40 line is 95% of the correction necessary to bring the sodium-24 interpolated energy into agreement with the known value. The results for a typical run of ten successive spectra, alternately potassium-40 and sodium-24, are given in Table I. The internal

TABLE II. Final summary.

K ⁴⁰ energy (keV)	Nonlinearity correction (keV)
1459.9±0.8	-10.9
1460.0±0.4	+2.3
1458.7±1.0	+0.9
1459.5±0.6	Simple average and probable error
1459.8±0.3	Weighted average
1459.7±1.0	Final result and assigned error

¹⁴ B. L. Robinson, NAS-NS 3107, Office of Technical Services, Department of Commerce, Washington, D. C., 1963 (unpublished), p. 67.

¹⁵ A. A. Bartlett, J. R. Keith, and W. D. King, Bull. Am. Phys. Soc. 8, 482 (1963).

¹⁶ G. Murray, R. L. Graham, and J. S. Geiger, Bull. Am. Phys. Soc. 7, 72 (1962).

probable error given there is the rms fluctuation of the five values about the average. For this particular experiment, the sodium-24 energy obtained by linear interpolation is about 11 keV too high, and the potassium-40 interpolated energy was corrected accordingly.

A small additional correction was made to account for the fact that the Compton distribution due to the 1836-keV line of yttrium-88 is slightly peaked in the region of the sodium-24 and potassium-40 full-energy peaks, with these two lines falling on opposite sides of the peak. Alternatively one may say that it was wrong to assume a constant contribution due to the Compton distribution, and that this correction would not be needed if the Compton distribution were more precisely described. This correction reduces the separation between the lines by about 0.5 keV.

FINAL RESULTS

Table II shows the results of three such experiments, performed under different conditions of phototube gain, amplifier gain, and ADC conversion. The nonlinearity corrections are different in each case, and yet the corrected results are in excellent agreement. The error as-

TABLE III. Dependence of final result on assumed values of standards.

Increase of 1 keV in	leads to
Na ²⁴ 1368.41	0.95 keV increase
Y ⁸⁸ 897.5	0.09 keV decrease
Y ⁸⁸ 1836.6	0.09 keV increase

signed to the final results is several times the internal error, and we believe that it covers any residual systematic error.

Table III shows how the final numerical result given here is to be corrected in case changes are made in the values adopted for the energies of the sodium-24 and yttrium-88 lines.

CONCLUSION

The energy of the potassium-40 gamma ray has been found to be 1459.7 ± 1.0 keV. As a consequence, it is clear that the discrepancy in the argon-potassium-40 mass difference⁷ does not lie in an erroneously low energy for the potassium-40 gamma ray.

ACKNOWLEDGMENTS

H. Jarosch of the Department of Applied Mathematics of the Weizmann Institute of Science advised and assisted in the initial programming of this work for the WEGEMATIC computer. Eric Williams of the Center for Documentation and Communications Research, Western Reserve University, rewrote the program for the General Electric 225 computer and ran the analyses

of the final data. Their contributions to this work are gratefully acknowledged.

APPENDIX

One hundred grams of potassium chloride enriched in potassium-40 was obtained on loan from the Stable Isotopes Division of Oak Ridge National Laboratory. This material had been produced by a long-term irradiation of potassium nitrate in the Materials Testing Reactor in order to enrich its potassium-40 content tenfold by radiative neutron capture in potassium-39. Following the irradiation, the sample had been converted to the chloride and it was being held for feed material for the electromagnetic-separation process.

The sample contained a readily perceptible amount of cesium-134, the only known long-lived activity of the

alkali metals which can be produced by neutron capture. The amount of cesium-134 corresponded to the activation of a cesium impurity of $10^{-8 \pm 0.5}$ parts per million. The radiocesium was removed by the addition of cesium-chloride carrier, followed by the precipitation of cesium silicotungstate at 0°C . Two repetitions of this process sufficed to reduce the cesium activity to an imperceptible level.

Following the removal of the cesium, the sample exhibited a residual activity which was sufficient to reduce significantly the "peak-to-valley" ratio in the scintillation spectrum, as compared to that from a comparable weight of natural potassium chloride. In the absence of any definite clues to the identity of the residual activity, no further work was done on this sample and it was returned to Oak Ridge.

Measurement of the Plane Polarization of Gamma Radiation*

JAMES N. HAAG†

*Lawrence Radiation Laboratory and Department of Chemistry,
University of California, Berkeley, California*

(Received 24 July 1963; revised manuscript received 23 January 1964)

A simplified experimental technique, utilizing Compton scattering as the analyzing process, has been developed for the measurement of the plane polarization of gamma radiation emitted from oriented nuclei. Nuclei of Ce^{139} and Ce^{141} were aligned in a neodymium ethylsulfate lattice at low temperatures by the magnetic-hyperfine-structure method. The plane polarization of the emitted gamma radiation was measured as a function of temperature. The measured polarization of this radiation was found to be consistent with published results.

I. INTRODUCTION

A PLANE-POLARIZED quantum has associated with it an electric-field vector perpendicular to its direction of propagation and in the plane of polarization whose orientation is constant in space. This quantum inelastically interacts with an electron so that the quantum is preferentially scattered into a plane perpendicular to its electric field vector. An analogous situation is a dipole antenna which radiates preferentially in a plane perpendicular to its length. The Compton scattering of gamma radiation is therefore a polarization-sensitive process. A comprehensive review article on gamma-ray polarization and its detection is that by Fagg and Hanna.¹

The plane polarization of gamma radiation emitted from oriented nuclei is ordinarily measured by using a polarimeter based on the one of Metzger and Deutsch.²

This type of polarimeter basically consists of a scattering crystal (usually an organic scintillator) of low gamma-ray detection efficiency so that the impinging quantum is scattered only once, and two detection crystals [usually $\text{NaI}(\text{Tl})$ scintillators] of high gamma-ray detection efficiency and situated perpendicular to each other. The inherent limitation of this type of polarimeter is its over-all detection efficiency, since a majority of the quanta pass through the scattering crystal without interacting. This limitation is partially overcome with this new technique by utilizing the gamma-ray source *itself* as the scattering medium, and thus eliminating the need of a scattering crystal.

The plane polarization of the 166-keV gamma ray from the decay of Ce^{139} and the 142-keV gamma ray from the decay of Ce^{141} were measured as functions of temperature, using a Metzger-Deutsch type of polarimeter. These results have been published.³ Then the plane polarizations of these gamma rays were measured as functions of temperature utilizing the new

* Work performed under the auspices of the U. S. Atomic Energy Commission.

† Present address: Department of Physics, Purdue University, Indianapolis, Indiana.

¹ L. W. Fagg and S. S. Hanna, *Rev. Mod. Phys.* **31**, 711 (1959).

² F. Metzger and M. Deutsch, *Phys. Rev.* **78**, 551 (1950).

³ J. N. Haag, D. A. Shirley, and D. H. Templeton, *Phys. Rev.* **129**, 1601 (1963).