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Masses of the Stable Chlorine Isotopes*

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The mass differences between the components of several doublets which involve the stable chlorine isotopes have been measured in a large mass spectrometer operating at a resolving power, at the base of the peaks, of 10^5 or better. These measurements lead to the following values of atomic mass, expressed as mass excesses on the $C^{12}=12$ u scale: $Cl^{37}=-34\,099.73\pm0.95$ μu; $Cl^{35}=-31\,148.62\pm0.68$ μu; $Cl^{37}-Cl^{35}=-2951.11\pm0.59$ μu.

INTRODUCTION

A PRECISE value for the mass difference $Cl^{37}-Cl^{35}$ is important in order to obtain accurate values for the binding energies of two nucleons from the series of measurements of the types $X^{A+2}Cl^{35}-X^{A}Cl^{37}$ and $X^{A+2}Cl^{35}-Y^{A}Cl^{37}$ reported by Duckworth and his associates.[1–8] These measurements have been used to study the systematics of nucleon binding.

Precise values of the atomic masses of Cl^{37} and Cl^{35} referred to C^{12} are useful as secondary standards of mass.

For the crucial mass difference, $Cl^{37}-Cl^{35}$, the best value available has been that obtainable from the mass table of Everling, König, Mattauch, and Wapstra.[9] The tabular value for Cl^{35} arises in the usual way from the general least-squares adjustment of many experimental data, some of them mass-spectroscopic doublet separations, some of them beta-decay energies, some of them nuclear-reaction energies.[10] But the tabular value for Cl^{37} is different. Because the links between Cl^{37} and nearby parts of the mass table are weak due to the relatively high errors involved, the tabular value for Cl^{37} depends almost entirely on two mass-spectroscopic determinations. These two determinations[11,12] are both of the doublet $C^{12}{}_3H^1-Cl^{37}$. Unfortunately, they are in disagreement by 8 times the sum of the stated errors. Before using any mass-spectroscopic results as input data for their least-squares adjustment the creators of the mass table increased the original errors by multiplying them by different "consistency factors."[10] In the case of the doublet $C^{12}{}_3H^1-Cl^{37}$ the stated error of the Minnesota measurement[11] was multiplied by 2.65 and the stated error of the Brookhaven measurement[12] was multiplied by 3.58. The disagreement is 2.6 times the sum of these increased errors. Thus the mass-table value for Cl^{37} is essentially a weighted compromise between two disagreeing measurements.

The measurements described in this paper have been undertaken in the hope of resolving the Cl^{37} discrepancy, finding more precise values for both Cl^{37} and Cl^{35}, and finding particularly a more precise value for the $Cl^{37}-Cl^{35}$ difference. Of the seven different doublets measured, three containing only the Cl^{37} isotope lead directly to a new value for Cl^{37}, three containing only the Cl^{35} isotope lead directly to a new value for Cl^{35}, and two containing both isotopes lead directly to a new value for the difference $Cl^{37}-Cl^{35}$.

APPARATUS AND MEASUREMENT METHODS

The large mass spectrometer at Harvard University has been described, at various stages of its development, by Collins and Bainbridge,[13] by Bainbridge and Moreland,[14] and by Moreland.[15]

Refinements continue to be made. The precision manganin resistors in the voltage-division circuit used to supply the potentials to the plates of the electrostatic analyzer are now bathed in flowing warm air the temperature of which is controlled to ±0.1°C. The batteries supplying the 1400 V for this circuit have been isolated from building vibrations which sometimes caused a modulation of the field in the electrostatic analyzer. A reduction was also made in electrical pickup in the leads to the electrostatic analyzer which could affect differently the two doublet components. The amount by

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[12] L. G. Smith, Phys. Rev. **111**, 1606 (1958).

[13] T. L. Collins and K. T. Bainbridge, *Nuclear Masses and Their Determination* (Pergamon Press Ltd., London, 1957), p. 213.

[14] K. T. Bainbridge and P. E. Moreland, Jr., *Proceedings of the International Conference on Nuclidic Masses* (University of Toronto Press, Toronto, 1960), p. 460.

[15] Parker E. Moreland, Jr., Ph.D. thesis, Harvard University, 1962 (unpublished).

which the ion accelerating voltage is alternated during high-mass–low-mass switching can now be set more accurately as a result of the introduction of a potentiometer circuit (operating at +80 kV) for the measurement of the current in the accelerating-voltage resistor chain.

The geometrical adjustments, position and angular orientation of the electrostatic analyzer with respect to the magnet, have been fixed at those values which are observed to give first-order direction focusing and first-order energy focusing consistent with the ions traveling in the center of the electrostatic analyzer and emerging as a parallel beam. In the Harvard instrument the image slit has been situated at that point on the focal plane for which there is second-order direction focusing, i.e., $B_{\alpha^2}=0$. (In the language of Hintenberger and König,[16] $B_{11}=0$.) The other aberration coefficients have been measured as $B_{\alpha\beta}=-5.2\pm0.5$ and $B_{\beta^2}=-1.4\pm0.2$, values consistent with the corresponding ones calculated from the Hintenberger and König expressions, $B_{12}=-6.1$ and $B_{22}=-1.41$.

Before a measurement is made the instrument is brought into energy focus and direction focus. Energy focusing occurs at the minimum in the curve of ion destination (in the direction perpendicular to the ion beam) versus ion energy and, in the oscilloscope display, at the corresponding minimum in the dependence of the position of the peak on ion accelerating voltage. The test for energy focus begins with the two component peaks of the doublet brought into superposition by a correct adjustment of ΔV, the amount by which the voltages on the plates of the electrostatic analyzer alternate. Adding to the accelerating voltage an amount $\Delta\phi$ during, say, the high-mass display time, and subtracting the same amount $\Delta\phi$ during the low-mass display time causes a displacement of both peaks. If these displacements are equal, i.e., if the peaks remain superimposed, the energy focus is correct. (In this test a typical $\Delta\phi$ value $=80$ V $=10^{-3}\times$ the accelerating voltage.)

To bring the instrument into direction focus the beam divergence is increased beyond its usual $\alpha=\pm0.3\times10^{-3}$ rad and l_e, the distance of the object from the electrostatic analyzer, is adjusted to give minimum peak width. The existence of the $\alpha\beta$ term (i.e., $B_{\alpha\beta}\neq0$) makes it important that adjustment for energy focus *precede* the adjustment for direction focus. Any change in energy *after* the direction focus adjustment introduces a first-power dependence of ion path on initial ion direction, i.e., the instrument is effectively no longer in first-order direction focus.

Immediately after each day's run the precision resistors in the circuit providing the voltages for the electrostatic analyzer are calibrated. The procedure involves a Julie Research Laboratories voltage divider of the Kelvin-Varley type which has been checked as having better than its claimed accuracy of 1×10^{-6}. The results of the calibration procedure cause the raw $\Delta m/m$ values, as read directly from the precision resistor circuit, to be modified in two ways, (1) by addition of a number, approximately $(10\pm1)\times10^{-8}$, (2) by multiplication by a number, approximately $1\pm(2\times10^{-6})$. Thus all $\Delta m/m$ measurements have an error due to resistor calibration of at least the $\pm1\times10^{-8}$ arising from the first factor. The error in $\Delta m/m$ contributed by the second factor is significant in the case of wide doublets (e.g., if $\Delta m/m=10^{-2}$, the error in $\Delta m/m=2\times10^{-8}$) but is trivial in the case of close doublets (e.g., if $\Delta m/m=10^{-4}$, the error in $\Delta m/m=2\times10^{-10}$). (These errors may be compared with the error in $\Delta m/m$ associated with the scatter of the individual observations which is about 1×10^{-8} when the resolving power is a typical 1×10^5 at the base of the peaks.)

About half the measurements reported in this paper have been made with the now traditional method of peak matching which involves visual observation of an oscilloscope display.[12] In our case the magnetic-field modulation is sinusoidal, usually 60 cps with the switching between high- and low-mass components of the doublet occurring every 1/60 sec. Blanking the oscilloscope trace during the first half of each cycle prevents observation of the peak while switching transients are still present. During a measurement which consists of 16 observations the ion-source conditions are usually changed 4 times and after each change the energy focus is checked and, if necessary, adjusted. The 16 observations involve the 16 possible arrangements of 4 two-position switches. The factors controlled by the switches are (1) whether the high-mass peak or the low-mass peak shall appear as a double-dotted line bracketing the other peak, (2) whether left-to-right on the oscilloscope shall mean increasing or decreasing magnetic field, (3) whether on the visible half of the sweep cycle the spot shall move left-to-right or right-to-left, (4) the phase of the 60 cps magnetic-field modulation relative to the 60 cps line.

The other measurements have been made with a newly developed method of peak matching. It involves a phase-sensitive amplifier to indicate as meter readings where, relative to the point of zero magnetic-field modulation, the high-mass and low-mass peaks lie. In this system the field modulation is approximately 1000 cps and the high-mass–low-mass switching currently occurs at 20 cps. An incidental advantage of this system is that it is possible to feed into the magnet signals which lock the field to the low-mass peak, to the high-mass, or to an average. We intend to give later a detailed description of this new method of peak matching.

A "measurement" in the case of peak matching by this new technique consists sometimes of 24 observations but more usually of 8 observations which represent different choices of signal phases and different assignment of channels to low-mass information and high-mass information.

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[16] H. Hintenberger and L. A. König, *Advances in Mass Spectrometry* (Pergamon Press Ltd., London, 1959), p. 16.

TABLE I. Measurements leading directly to the mass of Cl^{37}.

Doublet	Mass No. at which doublet occurs	No. of measurements	Δm (μu)	Components of error due to: Calibration factor	Scatter	Cl^{37} mass (expressed as mass excess in μu)	Components of error due to: Calibration factor	Scatter and standards
C_3H–Cl^{37}	37	3	41 924.73±1.09	0.84	0.69	−34 099.54±1.09	0.84	0.70
C_3H_3–DCl^{37}	39	6	43 473.27±1.33	0.87	1.00	−34 099.92±1.36	0.87	1.03
C_4H_6–CHD_2Cl^{37}	54	5	45 020.96±1.14	0.90	0.70	−34 099.45±1.23	0.90	0.84
					Weighted average=	−34 099.67±1.08	0.88	0.56

RESULTS

Mass differences have been measured for 8 types of doublets containing one or both of the stable chlorine isotopes. Of the three doublets which lead directly to the mass of Cl^{37}, one ($C_4H_6-CHD_2Cl^{37}$) does not contain molecular fragments. Two of the three doublets which lead directly to the mass of Cl^{35} ($C_6H_{12}-CH_2Cl^{35}{}_2$, $C_5H_8O-CH_2Cl^{35}{}_2$) also contain no fragment ions. And of the two doublets which lead directly to the mass difference $Cl^{37}-Cl^{35}$, one ($CHD_2Cl^{35}-CH_3Cl^{37}$) contains not only ions which are not molecular fragments but ions which are also chemically identical.

Each doublet has been measured at least 3 and as many as 7 times and each measurement, as explained earlier, involves 8 to 24 observations. The measurements stretched over 5 months and in only a few cases did more than one measurement of a particular doublet occur on the same day.

Each measurement of $\Delta m/m$ has associated with it an error which arises partly from the resistor calibration. The several measurements of each doublet have been weighted equally and averaged. Each average has associated with it its *external* error (arising from the scatter of the several measurements) rather than its *internal* error which was invariably smaller.

All Δm values have been multiplied by a calibration factor = 1.000 103±20 which was indicated by measurements of unit mass differences. To establish this calibration factor 10 measurements were made of the mass 54−53 difference in the doublet $C_4H_6-C_3H_3N$ and 6 measurements were made of the mass 85−84 difference in the doublet $C_5H_{11}N-C_5H_8O$. (Neither of these doublets involves molecular fragments.) These measured values were compared with the known mass differences.[17] Although the average of 16 values of the ratio Δm (known)/Δm (measured) was 1.000 103±8 (where ±8 is the *external* error of the average) we have pessimistically assigned an uncertainty of ±20 to the calibration factor.

The results of the mass-difference measurements are given in Tables I, II, and III. Table I, for example, shows for each of three doublets an average Δm (which includes the calibration factor) and the Cl^{37} mass which results.[18] These three Cl^{37} values were weighted in proportion to the number of measurements involved and an average was taken. Separate account was kept of the component of each error which was associated with the calibration factor because these components of the errors are not independent and do not tend to cancel as random errors do. In Table III, as in Table I, the error components due to the calibration factor happen to be approximately equal and the weighting is made proportional to the number of measurements involved. In Table II, because the three doublet separations are quite different, the error components due to the calibration factor are quite different and it also happens that the error components which are treated as random (those due to scatter in the measurements and to uncertainties in the standards) are quite different too. The weighting factors have been chosen so as to minimize the total error in the final average.

TABLE II. Measurements leading directly to the mass of Cl^{35}.

Doublet	Mass No. at which doublet occurs	No. of measurements	Δm (μu)	Components of error due to: Calibration factor	Scatter	Cl^{35} mass (expressed as mass excess in μu)	Components of error due to: Calibration factor	Scatter and standards
C_6H_{12}–$CH_2Cl^{35}{}_2$	84	6	140 549.37±2.98	2.81	1.00	−31 148.74±1.54	1.41	0.64
C_3H–DCl^{35}	37	6	24 871.92±0.75	0.50	0.56	−31 148.95±0.87	0.50	0.57
C_5H_8O–$CH_2Cl^{35}{}_2$	84	7	104 153.75±3.45	2.08	2.76	−31 143.93±1.75	1.04	1.41
					Weighted average=	−31 148.68±0.75	0.43	0.62

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[17] The "known" mass differences were obtained from the Mass Table of Everling, König, Mattauch, and Wapstra. The uncertainties in these mass differences are less than 0.5 μu and are therefore negligible in establishing the calibration factor.

[18] Used in these calculations are the following mass excesses for the standard masses as given in the 1964 Atomic Mass Table by J. H. E. Mattauch, W. Thiele, and A. H. Wapstra (to be published): $H^1 = 7\,825.19 \pm 0.08$ μu; $O^{16} = -5\,084.98 \pm 0.28$ μu; $D^2 = 14\,102.22 \pm 0.12$ μu.

TABLE III. Measurements leading directly to the mass difference, Cl^{37}–Cl^{35}.

Doublet	Mass No. at which doublet occurs	No. of measurements	Δm (μu)	Components of error due to: Calibration factor	Scatter	Cl^{37}–Cl^{35} mass difference (expressed as mass excess in μu)	Components of error due to: Calibration factor	Scatter and standards
DCl^{35}–Cl^{37}	37	3	17 052.95±1.02	0.34	0.96	−2 950.73±1.03	0.34	0.97
CHD_2Cl^{35}–CH_3Cl^{37}	52	7	15 505.41±0.71	0.31	0.64	−2 951.35±0.76	0.31	0.69
						Weighted average = −2 951.17±0.65	0.32	0.57

The three directly obtained values for Cl^{37}, Cl^{35}, and $Cl^{37}-Cl^{35}$ can be combined two at a time to give three indirect values for Cl^{37}, Cl^{35}, and $Cl^{37}-Cl^{35}$. For example, the difference between the direct value for Cl^{37} and the direct value for Cl^{35} is the indirect value for $Cl^{37}-Cl^{35}$. Expressed as mass excess this indirect value $= -2\ 950.99\pm0.94$ μu, and can be compared with the direct value given in Table III, $-2\ 951.17\pm0.65$ μu. The agreement shows evidence of good luck. A weighted average of the direct and indirect values $= -2\ 951.11 \pm0.59$. Similar weighted averages for Cl^{37} and Cl^{35} are given in Table IV.[19]

TABLE IV. Adjusted mass values.

	Mass or mass difference (expressed as mass excess in μu)
Cl^{37}	−34 099.73±0.95
Cl^{35}	−31 148.62±0.68
$Cl^{37}-Cl^{35}$	2 951.11±0.59

DISCUSSION

The 1960 Mass Table[9] value for Cl^{37} was derived essentially from two disagreeing experimental values for the doublet C_3H-Cl^{37}. These values are 41 921.3±1.2 μu (Minnesota[11]) and 41 938.6±0.98 μu (Brookhaven[12]). Our value for this doublet, based on only three measurements, is 41 924.73±1.09 μu. The Cl^{37} masses derived from the Minnesota and Brookhaven measurements are given in Fig. 1 along with the 1960 Mass Table value and our new value from Table IV. Our value is close to the Minnesota value but in obvious disagreement with the Brookhaven value.

Just after our measurements were completed we received from Professor Mattauch a copy of a new 1964 Atomic Mass Table.[20] In the absence of new direct mass-spectroscopic measurements of the chlorine isotopes, Mattauch, Thiele, and Wapstra[21] were forced to derive new masses for Cl^{37} and Cl^{35} by means of a general adjustment which made use of a combination of the Hamilton doublets[1–8] with new Minnesota doublets.[22,23] The new 1964 Mass Table value for Cl^{37} is given also in Fig. 1 and is seen to be in complete agreement with our new value.

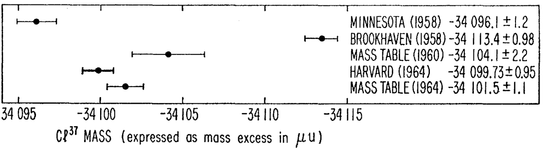

FIG. 1. Comparison of Cl^{37} mass values.

One of our doublets containing Cl^{35} can be compared directly with an earlier doublet measured at Minnesota.[11] Our measurements of the doublet $C_6H_{12}-CH_2Cl^{35}{}_2$ give for the mass difference $C_5H_{10}-Cl^{35}{}_2$ 140 549.37±2.98 μu. At Minnesota the doublet $C_5H_{10}-Cl^{35}{}_2$ was measured directly giving 140 540.3±3.4 μu for the separation. The Cl^{35} mass derived from this Minnesota measurement, the Cl^{35} masses from the 1960 and 1964 Mass Tables, and our new value from Table IV are compared in Fig. 2. Once again the 1964 Mass Table is in complete agreement with our new value.

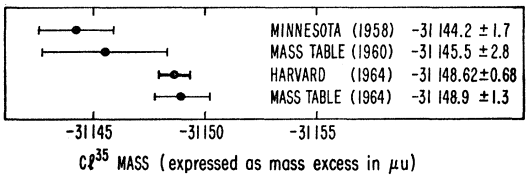

FIG. 2. Comparison of Cl^{35} mass values.

In Fig. 3 are comparisons of values for the $Cl^{37}-Cl^{35}$ mass difference. Our value is from Table IV. The others are differences between the corresponding values in Figs.

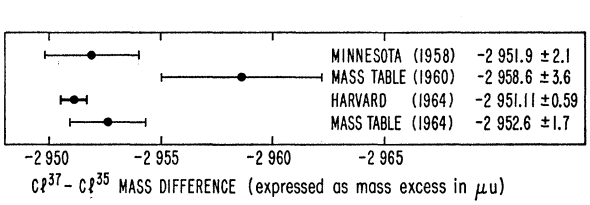

FIG. 3. Comparison of Cl^{37}-Cl^{35} mass-difference values.

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[19] Because the direct and indirect values happen to have similar dependent components of error, the weighting factors have been chosen inversely proportional to the squares of the independent components of error. In this case the weighted-average procedure is identical to a least-squares adjustment.

[20] J. H. E. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. (to be published).

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1 and 2. Our new value is significantly different from and significantly more precise than the best value previously available, i.e., from the 1960 Mass Table. Agreement with the 1964 Mass Table gives some confidence in its accuracy.

Note added in proof. Since our measurements were completed, new experimental values for the chlorine isotopes have been reported from Minnesota [J. L. Benson and W. H. Johnson, Jr., Phys. Rev. Letters **13**, 724 (1964)]. Expressed in terms of mass excesses, so that they can be compared easily with the earlier values in Figs. 1 to 3, these new Minnesota values are: $Cl^{37} = -34\,096.7 \pm 0.6\ \mu u$, $Cl^{35} = -31\,146.4 \pm 0.6\ \mu u$, $Cl^{37} - Cl^{35} = -2\,950.3 \pm 0.6\ \mu u$.

ACKNOWLEDGMENT

We are grateful to Professor Mattauch for sending us a preprint of the 1964 Atomic Mass Table.

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Energy Levels of Pb^{210} from Shell Theory*

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The present calculations for Pb^{210} have been based upon the double-closed-shell-core model with harmonic-oscillator wave functions. All two-neutron configurations of the $2g_{9/2}$, $1i_{11/2}$, $1j_{15/2}$, $3d_{5/2}$, $4s_{1/2}$, $2g_{7/2}$, $3d_{3/2}$ main shell are included. A singlet-even Gaussian effective interaction with parameters determined by low-energy proton-proton scattering and zero triplet-odd effective interaction give, within 50 keV, the binding energy of the two outer nucleons determined from experimental data and the energies of four excited states which have been experimentally identified. An empirical relation between matrix elements for slightly different parameters is described. It indicates that the interaction and the harmonic-oscillator parameter ν used here also give the general agreement with experiment which had been obtained for Pb^{206} with similar parameters. Calculations with a Yukawa singlet-even interaction and with additional attractive and repulsive central and central+tensor triplet-odd effective interactions were made for Pb^{210}; they all lead to somewhat worse agreement. None of these interactions is, however, definitely excluded by the present data. Level schemes, tables of level energies and of some wave functions, and detailed formulas for the calculation of γ-ray transition probabilities are given. A model for the 2.15-MeV $\frac{1}{2}^-$ state of Pb^{209} is also discussed.

I. BACKGROUND OF THE CALCULATION

In recent years calculations have indicated that energies of low states of nuclei near a double-closed-shell-core nucleus relative to the energy of such a nucleus can often be obtained from a shell-theoretical description with one or two single-particle states and matrix elements of the type

$$\mathfrak{M}(j_1j_2,j_1'j_2',J) = \langle j_1j_2J \,|\, V(1,2) \,|\, j_1'j_2'J \rangle. \quad (1)$$

Each single-particle state is described by the quantum numbers n_i, l_i, j_i; the first two numbers have been omitted in (1). In these investigations[1] by the "method of effective interactions," only a few matrix elements (1) appear; they are determined by the best fit to a large number of data. Often the fit of this theoretical model is excellent. It suggests that the detailed physical framework of the shell theory may have considerable validity at least in some regions of the periodic system.

Already long before enough data had become available for these investigations, *calculations* of the matrix elements (1) were made with simple assumptions for the two-nucleon interaction $V(1,2)$. These matrix elements are then used in larger calculations, which often take into account a considerable number of single-particle states. Nevertheless, there is usually less accurate agreement between such calculations and experiment than with the method of effective interactions.

A very striking exception, however, are the calculations by Kearsley[2] and True and Ford[3] for the nucleus Pb^{206}. There is now at least some indication from experimental data concerning the spins and parities of 19 excited states of this nucleus.[4] Three of these are tentatively identified as due to core excitation. Their excitation energies are approximately equal to those of probably corresponding excited states in the nucleus Pb^{208}. The measured energies of the 16 remaining excited states differ by an average of 2.7% from energies calculated with a singlet-even interaction between the outer nucleons.[5] Calculations have also been made for

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* Part of this work was done under the auspices of the U. S. Atomic Energy Commission. Calculations were made at the Computer Center, University of California, Berkeley.

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[2] M. J. Kearsley, Nucl. Phys. 4, 157 (1957).

[3] W. W. True and K. W. Ford, Phys. Rev. **109**, 1675 (1958).

[4] *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C.), NRC 61-4-110 to 126.

[5] W. W. True and K. W. Ford, Ref. 3, Table VIII.