Evidence from Nuclear Masses on Proposed Closed Shells at 20 Nucleons*

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Combining microwave measurements of sulfur and chlorine masses with mass spectroscopic and nuclear reaction information, the effect of nuclear shell structure on masses in the region of 20 neutrons or protons is investigated. It is found that except for Ca^{40}, nuclei with 20 neutrons or protons do not show any special stability.

THERE are various types of evidence[1–4] that neutron and proton numbers N and Z of 2, 8, 20, 50, 82 and 126 form particularly stable nuclei. These numbers can be associated with the completion of shells of the quantum levels of a single particle in a potential well.[5–7] Such a single particle model has been used with considerable success to correlate nuclear moments, isomerism, and β-decay. However, there has as yet been very little information about nuclear masses at N or Z of 20, 50, or 82. Hence, the most direct test for closed shells having extra stability has not yet been made.

Combining the recent microwave measurements of masses S^{36}, S^{34}, Cl^{36}, and Cl^{37} with mass spectroscopic and nuclear transmutation data, one can determine the masses of a large number of nuclei in the region of $N=20$ or $Z=20$. It is, therefore, feasible to test the expected variation in proton or neutron binding energy near the supposed closed shell at 20 nucleons. This is most conveniently done by comparison of the experimentally determined masses with those calculated from a semi-empirical formula which allows for most of the known sources of mass variation except shell structure. Such a formula has been given by Bohr and Wheeler[8, 9] as:

$$M_{AZ}=A-0.00081Z-0.00611A+0.014A^{\frac{2}{3}}+0.083(A/2-Z)^2/A+0.000627Z^2A^{-\frac{1}{3}}+\lambda,$$

where A = mass number, Z = nuclear charge, $\lambda=0$ for A odd, $\lambda=-0.036A^{-\frac{3}{4}}$ for A even, Z even, $\lambda=0.036A^{-\frac{3}{4}}$ for A odd, Z odd.

This equation thus takes into account the normal odd-even fluctuations of masses. Although in general this expression does not agree with the experimentally measured masses to high accuracy, nevertheless the difference between the experimental and calculated masses plotted as a function of N or Z should give a relatively smooth curve. Any marked deviation from this curve, i.e., a sudden change in slope, might indicate the effects of shell structure.

Figures 1 and 2 show a plot of the difference between

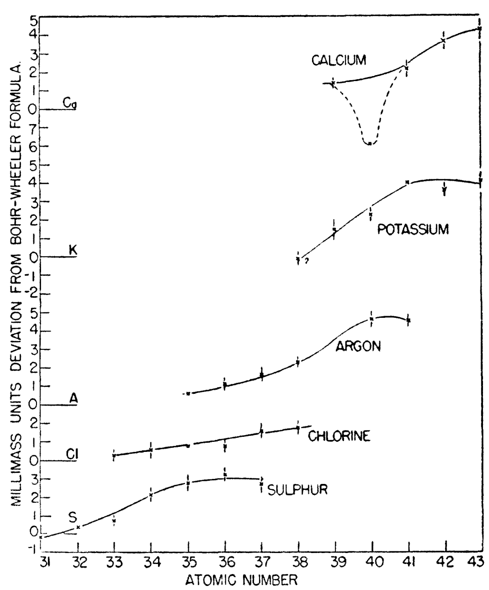

FIG. 1. Variation of nuclear mass with fixed Z and varying neutron number.

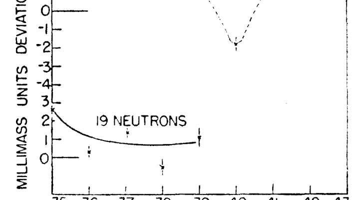

FIG. 2. Variation of nuclear mass with fixed neutron number and varying Z.

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TABLE I. Mass differences calculated from microwave data and nuclear reactions.

$H^1=1.008128$, $D^2=2.014718$, $n=1.008938$, $\alpha=4.003880$ mass units

Nuclei	Calculated mass difference	Reactions used	Reference
$P^{32}-P^{31}$	1.00028±40	dp	a
$S^{31}-P^{31}$	0.00519±17	β^+	b
$P^{32}-S^{32}$	0.00179±20	β^-	c
$S^{34}-P^{31}$	2.99542±40	αp	d
$P^{34}-S^{34}$	0.000547±20	β^-	e
$S^{33}-S^{32}$	0.99965±05	dp	f
	0.99977±30	mic.	g
$S^{33}-S^{34}$	0.99725±05	dp	f
	(0.99709±15)	(mic.)	
$S^{34}-S^{36}$	2.00054±30	mic.	h
$Cl^{35}-S^{32}$	2.99800±40	αp	i
$Cl^{35}-S^{33}$	1.99893±40	$d\alpha$	j
$S^{35}-Cl^{35}$	0.000179±03	β^-	k
$Cl^{33}-S^{33}$	0.00549±07	β^+	b
$Cl^{34}-S^{34}$	0.00656±10	β^+	l
$Cl^{35}-Cl^{37}$	1.99751±14	mic.	m
$Cl^{35}-Cl^{36}$	0.99988±30	dp	n
	1.00017±40	mic.	o
$S^{37}-Cl^{37}$	0.00461±20	β^-	e
$Cl^{38}-Cl^{37}$	1.00228±30	dp	p
$A^{35}-Cl^{35}$	0.00580±30	β^+	b
$Cl^{36}-A^{36}$	0.000768±.000006	β^-	k
$A^{37}-A^{36}$	0.99961±05	dp	q
$A^{38}-Cl^{35}$	2.99443±40	αp	r
$Cl^{38}-A^{38}$	0.00558±20	β^-	s
$K^{38}-A^{38}$	0.00641±20	β^+ ($\gamma+\beta^+$-ray in cascade probably)	t
$A^{41}-A^{40}$	1.00249±05	dp	u
$A^{41}-K^{41}$	0.00274±20	β^-	v
$K^{40}-A^{40}$	0.0017±20	pn	w
	≥0.00167±10	K-capture	x
$K^{40}-K^{39}$	1.00070±07	dp	y
$Ca^{39}-K^{39}$	0.00619±30	theoretical	
$K^{40}-Ca^{40}$	0.00145±05	β^-	aa
	0.00148±03	β^-	bb
$Ca^{41}-Ca^{40}$	0.99997±05	dp	cc
$Ca^{41}-K^{41}$	0.00047±07	pn	dd
$K^{39}-Ca^{42}$	2.99669±40	αp	ee
$K^{42}-Ca^{42}$	0.00383±20	β^-	v
$K^{43}-Ca^{43}$	0.000868±03	β^-	ff
$Sc^{41}-Ca^{41}$	0.00630±20	β^+	b
$Sc^{43}-Ca^{43}$	0.00226±25	β^+	gg
$Ca^{40}-Sc^{43}$	3.00036±40	αp	ee
$K^{41}-K^{41}$	1.00109±10	αp	hh
$Ca^{40}-A^{40}$	0.00032±08	mass-spectros.	ii

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[cc] See reference 21.

the experimental and computed masses against A for various Z and N. Tables I and II summarize data employed for determination of the masses, the mass values, estimates of their probable errors, and the references. Figure 3 indicates the experiments which relate the various nuclear masses.

TABLE II. Atomic masses based on Table I.*

Standard: $S^{32}=31.98089$, $A^{40}=39.97516$

Nucleus	Mass, based on Table I	Comment	Masses computed from Bohr-Wheeler formula	Δ difference in m.m.u.
P^{31}	30.98239±40	Average of $P^{31}\alpha pS^{34}$ and $P^{31}dpP^{32}$ reactions	30.98218	0.21
P^{32}	31.98268±20		31.98315	−0.47
P^{34}	33.98326±25		33.98219	0.07
S^{31}	30.98758±50		30.98756	0.02
S^{32}	31.98089	Standard all nuclei from P^{31} to A^{40} calculated with respect to S^{32}	31.98051	0.38
S^{33}	32.98058±10		32.98011	0.47
S^{34}	33.97780±20	Average of microwave and $S^{32}-dp-S^{33}$ reaction	33.97566	2.14
S^{35}	34.97907±45		34.97649	2.58
S^{36}	35.97834±40		35.97510	3.24
S^{37}	36.98111±50		36.97866	2.45
Cl^{33}	32.98607±10		32.98575	0.32
Cl^{34}	33.98436±25		33.98391	0.45
Cl^{35}	34.97889±40	Average of $S^{32}-\alpha p-Cl^{35}$ and $S^{33}-d\alpha-Cl^{35}$	34.97817	0.72
Cl^{36}	35.97892±50	Average of $Cl^{35}dpCl^{36}$ and microwave data	35.97855	+0.37
Cl^{37}	36.97640±50		36.97503	1.37
Cl^{38}	37.97868±55		37.97728	1.40
A^{35}	34.98468±50		34.98406	0.63
A^{36}	35.97822±55		35.97717	1.05
A^{38}	37.97320±60	Average of $Cl^{35}\alpha pA^{38}$ and $Cl^{38}\beta^-A^{38}$ reactions	37.97138	1.82
A^{40}	39.97516±26	Standard, reevaluated using references 10, 11 (Mattauch)	39.97020	4.96
A^{41}	40.97765±05		40.97293	4.72
K^{38}	37.97905±70	Assuming β^+ and γ are in cascade	37.98035	−1.30
K^{39}	38.97613±30		38.97460	+1.53
K^{40}	39.97683±10		39.97446	2.37
K^{41}	40.97491±25		40.97075	4.16
K^{42}	41.97600±30		41.97235	3.65
K^{43}	42.97433±65		42.97038	3.95
A^{37}	36.9783 ±60		35.97634	1.49
Ca^{39}	38.98232±40	Theoretical value using $\beta^+=4.8$ Mev.	38.98100	+1.32
Ca^{40}	39.97546±08		39.97701	−1.55
Ca^{41}	40.97543±12		40.97299	2.44
Ca^{42}	41.97217±35		41.96829	3.88
Ca^{43}	42.97346±55		42.96888	4.58
Sc^{41}	40.98173±35		40.97955	2.18
Sc^{43}	42.97572±50		42.97141	4.29

* Note that masses and errors given are not absolute values, but are relative to the masses chosen for S^{32} and A^{40}.

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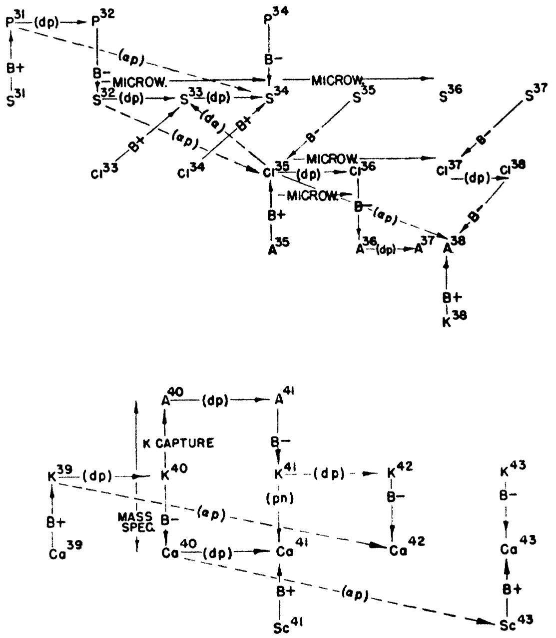

FIG. 3. Known connections between masses of certain nuclei.

II. DISCUSSION OF MASS DETERMINATIONS

All masses from P^{31} to A^{40} have been calculated with respect to the mass of $S^{32}=31.98089$. The masses from argon to scandium have been computed with respect to A^{40}. The mass of A^{40} has been taken as 39.97516 ± 0.00028 determined from the doublet[10] $(Ne-A/2) = 111.42\times10^{-4}\pm0.38$ and using[11] $Ne^{20}=19.99872 \pm 0.00013$. All reactions used for the determination of masses have been recalculated using Bainbridge's values[11] for the masses of H, n, D, and α. The mass values of a few nuclei require further explanations as given below.

S^{31}.—S^{31} is positron active and has been determined with respect to S^{32} through the mass of P^{31}, as is indicated in Fig. 3. The P^{31} mass obtained is 30.98238 ± 45 or 30.98240 ± 40 for the reaction paths involving S^{34} or P^{32} respectively. The S^{31} mass is obtained immediately from P^{31}.

Cl^{35}.—The mass of Cl^{35} can be determined from two different reactions as shown in Fig. 3. From the αp reaction $Cl^{35}=34.97889 \pm40$ and from the $d\alpha$-reaction $Cl^{35}=34.97888\pm40$. For the αp reactions Bethe's revised Q values have been used throughout.[12]

Cl^{37}.—The Cl^{37} mass is determined from the mass ratio of Cl^{35}/Cl^{37}. Unfortunately there are different values for this ratio from mass spectroscopy and microwave spectroscopy. A recent mass spectroscopic determination[13] of this ratio gives the value as 0.9459441 ± 65 (Aston[14] had found earlier the ratio as 0.9459806, but his experimental error is stated as ±300). This value disagrees with the values obtained from microwave measurements[15,16] in ICl and FCl which give 0.9459801 ± 50 and 0.9459775 ± 40 respectively. Isotopic frequencies for Cl^{35}, Cl^{36}, and Cl^{37} in the molecule ClCN have also been measured.[16a] With these and a knowledge of the mass difference $Cl^{35}-Cl^{36}$ (obtained from $Cl^{35}\,dp\,Cl^{36}$ reaction) one finds this ratio $Cl^{35}/Cl^{37}=0.9459906\pm120$ in agreement with other microwave results. Moreover, an evaluation of this ratio from transmutation data[17,18] leads to a value in essential agreement with microwave results. Hence the most recent mass spectroscopic value must be in error. Table III summarizes these results.

A^{38}.—A^{38} may be determined from Cl^{35} with an α,p reaction and from the beta-decay of Cl^{38}. The values are 37.97332 and 33.97322 ± 60 respectively.

K^{38}.—K^{38} gives off a positron of 2.53 Mev and a γ-ray of 2.15 Mev. We assume that these are in cascade.

Ca^{39}.—Ca^{39} belongs to a class of nuclei with $Z-N=1$ which are positron active. These "mirror" nuclei have been studied extensively; their half-lives and positron energies form a very regular and apparently well-understood series. The half-life of Ca^{39} has been measured recently[19] and is 1.06 sec. in good agreement with expectations from this series. Hence the predicted[20] but still unmeasured† value of 4.8 Mev for the positron energy seems to us very reliable for obtaining the mass difference $K^{39}-Ca^{39}$.

Ca^{40}.—The mass difference $Ca^{40}-A^{40}$ has recently been obtained as $(3.2\pm0.8)\times10^{-4}$ from mass spectroscopy[20a] and as $(2.7\pm2.1) \times10^{-4}$ from nuclear reactions.[21]

K^{43}, $Ca^{42,43}$, Sc^{43}, Ca^{42}.—The Ca^{42} mass has been determined from beta-decay of K^{42}. From the measurement by Sailor[21a] of the mass difference $K^{42}-K^{41}$ one obtains for the mass of $Ca^{42} =41.97217\pm35$. If the $K^{39}\,\alpha p\,Ca^{42}$ reaction reported by Pollard and Brasefield[22] is used, a value 41.97293 ± 50 results. This gives support to Bethe's suggestion[12] that the αp reaction measured does not represent a transition to the ground state. Similarly, it is not quite certain whether the observed $Ca^{40}\,\alpha p\,Sc^{43}$ reaction represents a transition to the ground state, and therefore, the

TABLE III. Value of Cl^{35}/Cl^{37} ratio.

Name	Method	Value
Aston[a]	Mass spectroscopy	0.9459806 ± 300
Okuda *et al.*[b]	Mass spectroscopy	$0.9459452\pm\ 65$
Townes *et al.*[c]	Microwave (ICl)	$0.9459801\pm\ 50$
Roberts *et al.*[d]	Microwave (FCl)	$0.9459775\pm\ 40$
Townes *et al.*[e]	Microwave (ClCN)	0.9459906 ± 120
Pollard[f]	Nuclear reaction	0.9459893 ± 110

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[b] See reference 13.
[c] See reference 15.
[d] See reference 16.
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masses of Ca^{43}, K^{43}, and Sc^{43} given in Table I may be somewhat too high.

III. DISCUSSION OF RESULTS

Figures 1 and 2 show that although there is considerable deviation of the measured masses from the Bohr-Wheeler formula, all deviations with the exception of the value for Ca^{40} lie on relatively smooth curves. The deviations from the curves are not generally larger than 0.5 Mev. There seems to be no abrupt change of mass or of slope of the curves of mass *versus* neutron number near S^{36}, Cl^{37}, A^{38}, or K^{39}, all of which are atoms with 20 neutrons. Similarly, there is no evidence of shell structure at 20 protons from the curves of mass *versus* proton number, except in the case of Ca^{40} which shows a striking deviation from the smooth curves of about 3.5 Mev. This deviation is far greater than the probable error of mass determinations or than the deviation of any other nucleus plotted.

The absence of any change in slope at 20 nucleons makes it rather questionable whether 20 nucleons should be regarded as the closing point of a major shell. The well-known stability of Ca^{40} seems not to be simply connected with the completion of a shell at 20 nucleons. This case indicates that some large deviations from the stability curve may be encountered for other nuclei which are not attributable to neutron or proton shells alone but depend on the combination of neutron and proton numbers. Perhaps the large mass spread of stable Ca isotopes is due to this exceptional stability of Ca^{40} and to a shell at 28 neutrons which makes Ca^{48} stable rather than to a general stability of 20 protons.

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On a Difference Equation Method in Cosmic-Ray Shower Theory*

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The recent results of Snyder and Bhabha-Chakrabarty for the cascade theory of cosmic-ray showers are shown to be derivable from a general approach involving the use of the Laplace and Mellin transforms, and a general and powerful method, due to Snyder, for solving the resulting difference equations. Boundary conditions are introduced in a natural and automatic way, and the accuracy of the solution is limited by the possible ways of evaluating the resulting triple complex integral.

I. INTRODUCTION

SNYDER[1] has recently obtained numerical results for the cascade theory of electron-photon showers which appear to be considerably more accurate than those of Bhabha and Chakrabarty.[2] It is the object of this paper to present a general method of solving the shower equations which yield both of the above-mentioned solutions, and which should be applicable to a number of other problems.

II. THEORY

Using Snyder's[1] notation, we write the diffusion equation for $P(E, t)$, the mean energy spectrum of electrons at depth t, and $\gamma(E, t)$, the mean energy spectrum of photons:

$$\frac{\partial P(E,t)}{\partial t}=\lim_{\delta\to 0}\left[\int_{E+\delta}^{\infty}P(E',t)R(E',E'-E)\frac{E'-E}{E'^2}dE' - P(E,t)\int_{\delta}^{E}R(E,E')\frac{E'dE'}{E^2}\right]+\beta\frac{\partial P(E,t)}{\partial E} + 2\int_{E}^{\infty}\gamma(E',t)R(E,E')\frac{dE'}{E'}, \quad (1)$$

$$\frac{\partial \gamma(E,t)}{\partial t}=\int_{E}^{\infty}P(E',t)R(E',E)\frac{EdE'}{E'^2} - \gamma(E,t)\int_{0}^{E}R(E',E)\frac{dE'}{E}. \quad (2)$$

In these equations $R(E', E)$ is a function which yields the elementary probabilities per unit path length of the pair-production and bremsstrahlung processes. In the case of high energies, the asymptotic form of R is that of a function homogeneous in E/E'; this is the only case dealt with in the present treatment.

* This work was started in 1940 (Ph.D. Thesis, University of Michigan, 1941). It was completed at the Brookhaven National Laboratory, under the auspices of the AEC.

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