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On-Line Mass-Spectrometric Measurement of the Masses of Neutron-Rich Sodium Isotopes

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(Received 29 January 1973)

Using an on-line mass spectrometer, we have measured directly, to an accuracy of 150 to 500 keV, the masses of $^{27-30}\text{Na}$ produced in the reaction of 24-GeV protons on uranium.

Light nuclei with a large excess of neutrons have been produced using the reaction of high-energy protons on heavy targets^{1,2} or in complex transfers from heavy-ion-induced reactions.³ In most of the cases only the existence (i.e., particle stability) of the more exotic nuclei is known. This information is of particular value if the calculated limits of nuclear stability can be tested this way. Some experiments also give other ground-state characteristics such as half-lives² or masses.⁴ The ground-state mass measurements reported to date are based on transfer reactions^{4,5} or β end-point measurements⁶ and appear to be limited to nuclei of $T_z \leq 3$ (for $Z \leq 15$).

In this paper we want to report for the first time direct mass measurements with an on-line mass spectrometer. These first results concern sodium isotopes (^{27}Na to ^{30}Na) produced in the reaction of 24-GeV protons on uranium at the CERN proton synchrotron. While ^{30}Na is already $T_z = 4$, there is a definite possibility to extend these measurements even further from stability.

The importance of this point lies in the fact that the mass calculations that serve to predict the stability of very neutron-rich nuclei all derive their parameters from a region of nuclei with measured masses near the valley of β stability. Very distant extrapolations can then lead to sizable errors as can be seen by observing that the predictions of different mass formulas⁷ can differ between themselves by several MeV for nuclei of $T_z \geq 3$. Hence the measurement of masses of nuclei far from stability can lead to a very severe test of the validity of mass formulas even when the measurements have an accuracy of only 200–500 keV. Measurements to this accura-

cy for sodium isotopes (about 10^{-5}) can be obtained with a spectrometer with a resolving power of only 500 [full width at half-maximum (FWHM)] by measuring the centroid of the peaks to an accuracy of $\frac{1}{2}\%$ of their widths.

On-line mass spectrometry had already been used to identify new isotopes and measure their half-lives.² The essence of the technique is the capture of energetic recoils from the reaction in heated graphite foils from which alkali elements diffuse very quickly to be ionized on a hot rhenium surface. A schematic of the experiment is shown in Fig. 1. For the rest of the discussion it is appropriate to think of the instrument as an ordinary mass spectrometer with a special surface-ionization ion source that is loaded with the short-lived nuclei of interest at regular time intervals (every 10 sec) by the interaction of intense ($\sim 10^{12}$) proton bursts with a target consisting of about 2 g/cm² of uranium. The resulting ions are accelerated with about 10 kV and analyzed by a slightly inhomogeneous ($n = 0.23$) magnetic prism ($\varphi = 90^\circ$, $r = 35$ cm). Two triplets of electrostatic quadrupole lenses are used to refocus the ions passing the exit slit of the spectrometer onto an electron multiplier located in a well shielded area and able to detect single ions.

The measurement of masses rests on the same theorem that is used in high-resolution mass measurements⁸: two ions A^+ and B^+ of masses M_A and M_B follow the same trajectory in the spectrometer if the magnetic field configuration remains constant and all electrostatic potentials are changed to satisfy the ratio

$$M_A/M_B = V_B/V_A. \quad (1)$$

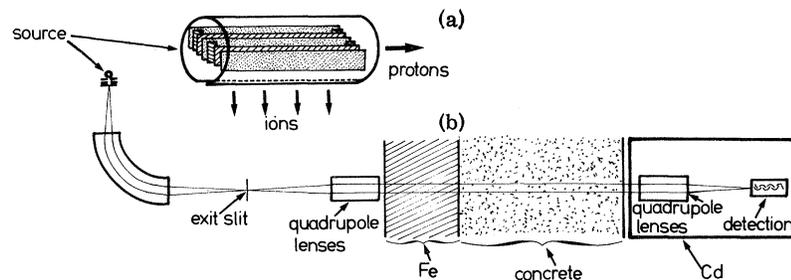


FIG. 1. (a) The target-ion source consists of an array of graphite foils coated with uranium and wrapped in a rhenium envelope. (b) Schematic layout of the experiment, in a plane perpendicular to the proton beam. The ions passing through the exit slit of the spectrometer are refocused by electrostatic lenses on an electron multiplier located in a well shielded area.

To measure the ratio of two masses with our instrument it is thus necessary to keep the field fixed and measure the accelerating potentials V_A and V_B corresponding to ions that follow the same trajectories to the exit slit of the spectrometer. To meet this condition, a calibrated triangular modulation is applied to the dc accelerating potential U in order to sweep the ions past the slit. From the peak in counting rate and the known modulation, an increment v is determined (see Fig. 2) so that one has $V_A = U_A + v_A$ and $V_B = U_B + v_B$.

During alternate pulses of the synchrotron (i.e., every 10 sec) data are recorded on masses A and B . U_A and U_B come from two physically distinct highly regulated (2×10^{-5}) power supplies and are switched by a system of relays. The dc potentials (6 to 10 kV) are measured each beam burst through a dividing bridge with a digital voltmeter accurate to 10^{-6} . To determine the increments v , the centroids of the peaks (both means and medians) are calculated during the experiment with a PDP-15 computer.

In order to account for systematic errors, Eq. (1) was modified by introducing the parameter δ such that $M_A/M_B = (V_B + \delta)/(V_A + \delta)$. The parameter δ probably accounts for many causes of systematic errors. Some of these are obvious like the constant 3-V dc heating of the surface ionization source which is not changed when jumping from mass A to mass B . Also, the contact potentials are certainly different in the two positions of the relay corresponding to mass A and mass B . There are, of course, other causes of systematic errors that still remain unknown to us because the method is so new.

It was thus felt indispensable to calibrate the instrument by measuring δ on pairs of known masses in actual experimental conditions. A

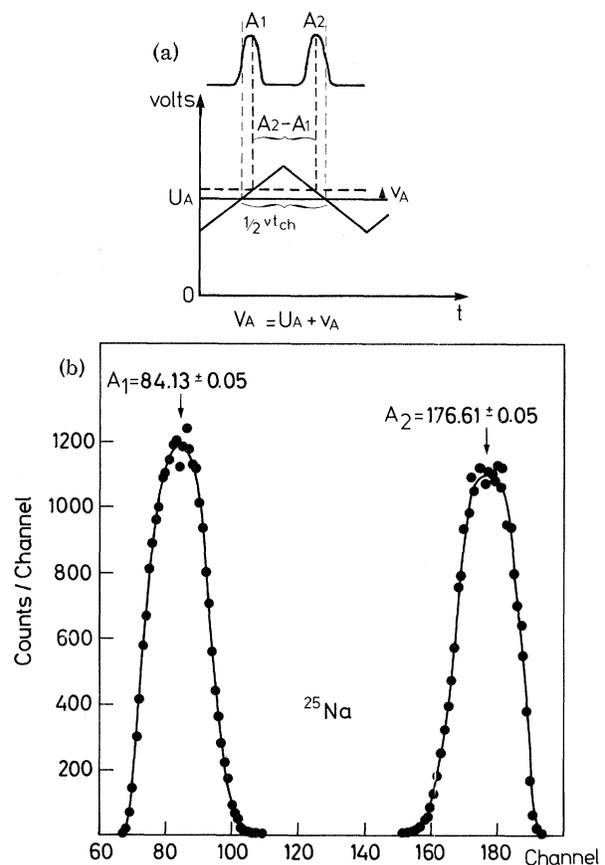


FIG. 2. (a) To determine the total accelerating potential V , a triangular modulation is added to the dc accelerating potential U . The increment is determined by comparing the difference of the centroids of the two peaks ($A_2 - A_1$) with one half of the wavelength of the modulation ($\frac{1}{2}v_{ch}$). On the alternate pulses of the proton synchrotron data are recorded on mass A (here shown) and mass B (not shown) by switching a system of relays. (b) In a typical case, the statistical accuracy in the determination of the center of gravity of the peaks can be better than 0.5% of the FWHM. No particular assumption has to be made on the regularity of the peak shape in as much as it remains constant with mass.

TABLE I. Atomic mass excesses ($M-A$) in keV. The numbers in parentheses are the number of measurements.

Reference mass	New mass				
	27	28	29	30	
^{25}Na	-9356	-5775±635 (2)	-1014±200 (9)	3105±495 (4)	
^{26}Na	-6854±30	-5881±145 (4)	-1619±475 (1)	2665±260 (6)	8195±685 (3)
^{27}Na	-5880±140		-2249±460 (2)		7686±1360 (1)
^{28}Na	-1260±170			2576±480 (2)	
^{29}Na	2730±210				8712±690 (2)
Best value		-5880±140	-1260±170	2730±210	8370±460

^aSee Ref. 9.

systematic study of Na isotopes with known masses⁹ showed δ values of the order of 8 V. While this value in itself is not unreasonable, its long-term variation during the time of the experiment was relatively large [$\sigma(\delta) = 2.4$ V] and thus it was decided to measure δ at least twice just before and just after each unknown mass measurement with two known masses differing by the same mass-number jump. For example, a measurement of $M(^{25}\text{Na})/M(^{27}\text{Na})$ would be calibrated before and after by $M(^{24}\text{Na})/M(^{26}\text{Na})$.

The results of the mass determinations are presented in Table I with their errors determined from the errors in the measurement of the voltages V_A and V_B and the uncertainty in the quantity δ . The errors in the voltage measurements come primarily from the counting statistics that determine the centroid of the peak. The additional error introduced by the digital voltmeter (DVM) measurement is certainly smaller and is overestimated by taking the dispersion in the DVM readings which reflect more the actual drifts of the power supply. These result in a broadening of the peak and thus affect the counting statistics. For the δ measurements which had good statistics, these unknown DVM measurement errors could not be neglected any more. Thus the δ errors were determined from the agreement of replicates and came to approximately one volt. A χ^2 value was calculated for the deviation of the 36 independent mass measurements represented in Table I from the four means and came to 26 for 32 degrees of freedom. The reproducibility of the mass measurements therefore agrees with what one would expect from our calculated er-

rors. It is also worth pointing out that our accuracy seems at present limited by counting statistics, rather than by systematic errors.

A comparison of our masses with both the original and the revised Garvey *et al.* mass calculations^{10,11} is shown in Table II. The predictions are lower than the measured masses of ^{29}Na and ^{30}Na . Also other mass formulas are low by 2 to 6 MeV. A parallel discrepancy for the revised Garvey *et al.* formula has been observed for other recently measured masses like ^{22}O ⁴ and ^{33}Si ,⁶ although ^{35}P ⁶ and ^{21}O ⁴ agree with the predictions and ^{29}Mg ⁵ and ^{25}Ne ⁶ go the other way. If this is a trend toward lower calculated masses it at first appears inconsistent with the fact that isotopes such as ^{11}Li , ^{14}B ,¹ ^{19}C ,¹² and ^{32}Na ² had been found experimentally bound when predicted marginally unbound by mass calculations.^{10,11} The contradiction may be only apparent since nuclear stability depends on neutron (or two-neutron) separation energies and not on total binding energies. In the case of sodium isotopes we can deduce neutron separation energies from our measurements and compare them with the calculated values. It is apparent from Table II for ^{29}Na and ^{30}Na that the calculated odd-even effect is significantly greater than the experimental one, the difference being smaller for the revised mass formula. This trend might account for the observed stability of ^{32}Na , ^{19}C , and ^{14}B since they are all odd-neutron nuclei. It is obvious that much more data are needed on the masses of nuclei far from stability before a general pattern emerges and definite conclusions can be drawn. We think that many results along these

TABLE II. Comparison with calculations.

Isotopes	T_z	Experimental	$M-A$ (MeV)		Experimental	E_n (MeV)	
			Garvey <i>et al.</i> revised ^a	Garvey <i>et al.</i> 1969 ^b		Garvey <i>et al.</i> revised ^a	Garvey <i>et al.</i> 1969 ^b
²⁷ Na	$\frac{5}{2}$	-5.88 ± 0.14	-5.98	-6.65	7.10 ± 0.14	7.10	7.10
²⁸ Na	3	-1.26 ± 0.17	-1.55	-3.38	3.45 ± 0.22	3.64	4.80
²⁹ Na	$\frac{7}{2}$	2.73 ± 0.21	0.86	-2.14	4.08 ± 0.27	5.66	6.84
³⁰ Na	4	8.37 ± 0.46	6.89	2.72	2.43 ± 0.51	2.04	3.21

^aSee Ref. 11.^bSee Ref. 10.

lines will be obtained in the future, using the technique that is described here for the first time.

It is a pleasure to acknowledge the excellent contributions of R. Ferreau, M. Jacotin, and J. F. Kepinski in building the mass spectrometer and their able assistance during the experiment at CERN. We thank G. Le Scornet for preparing the targets and assistance in data processing. We are grateful to Alan Ball and all the members of the group at CERN whose help was essential in installing the experiment and maintaining the complex protons beam transport.

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Nuclear Response Function

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(Received 23 May 1973)

A fast numerical method is described for calculating nuclear excitation properties with δ -type interactions. The method is applied to ²⁰⁸Pb, and the following properties of the nucleus are found: (i) The quadrupole strength has two main pieces, one a low state and one identifiable as the giant quadrupole; (ii) excitations with $L > 2$ do not seem to have high-energy collective parts; (iii) the giant dipole $L=1, T=1$ is too low unless the interaction has a strong momentum dependence.

There has been much interest recently¹⁻³ in Hartree-Fock calculations of nuclei using the parametrization of the nucleon interaction in terms of momentum and density-dependent δ functions, called Skyrme interactions.⁴ If the Hartree-Fock description of a ground state is reasonable, the random-