Nuclear charge radii of the even sulphur isotopes ³²S, ³⁴S, and ³⁶S and of ³¹P using muonic atoms

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Energies of muonic x rays of the Lyman series of the even sulphur isotopes ³²S, ³⁴S, and ³⁶S and of natural phosphorus have been determined with absolute precisions up to 23 parts/10⁶. Equivalent Barrett charge radii $R_{k,\alpha}$ have been deduced. Their differences between the sulphur isotopes amount to $\Delta R_{k,\alpha}({}^{34}S-{}^{32}S)=29.7(1.4)$ am and $\Delta R_{k,\alpha}({}^{36}S-{}^{34}S)=18.7(1.5)$ am. Combining these results with recent elastic electron scattering data, we obtain in a model-independent way $\Delta \langle r^2 \rangle^{1/2}({}^{34}S-{}^{32}S)=23.1(1.2)$ am and $\Delta \langle r^2 \rangle^{1/2}({}^{36}S-{}^{34}S)=13.0(1.2)$ am.

In order to extend our measurements of the systematic behavior of nuclear charge radii upon addition of a pair of neutrons,¹⁻³ we have determined the nuclear charge radii of the even sulphur isotopes ${}^{32}S$, ${}^{34}S$, and ${}^{36}S$ using the method of muonic x rays. The sulphur isotopes complete the $1d_{3/2}$ neutron shell up to the magic neutron number N=20. Their equivalent charge radii $R_{k,\alpha}$ and their differences $\Delta R_{k,\alpha}$ have been determined to an accuracy of better than 3 am and 1.5 am, respectively. Similar precision has been obtained for ³¹P, which was employed for reference purposes. The core polarization effect due to the addition of neutron pairs in the three even sulphur isotopes follows the trend of the even argon isotopes⁴ and the recently published ${}^{12}C$, ${}^{13}C$, and ${}^{14}C$ data.³ However, there is no decrease of the nuclear charge radius in going from ${}^{34}S$ to ${}^{36}S$ as indicated by the recent elastic electron scattering data of Rychel *et al.*,⁵ and as observed for several heavier isotope chains just below the N=28 and 50 neutron shell.^{2,6,7} Preliminary results of the present experiment have been reported at conferences.^{8,9}

The experiment has been performed at the $\mu E1$ channel of the SIN accelerator at Villigen, Switzerland. Setup and electronics were similar to former runs.¹⁰ A 50 cm³ truecoaxial Ge(Li) detector with a resolution (FWHM) of 1.6 keV at 516 keV and a 6.5 cm³ intrinsic planar Ge detector with a resolution of 1.2 keV at the same energy have been employed. The three sulphur targets consisted of 2 g of natural sulphur, 200 mg of enriched ³⁴S, and 150 mg of enriched ³⁶S. Table I gives the respective isotopic compositions. With each of the three sulphur targets, 2.5 g of natural phosphorus powder was simultaneously measured. In that way, the same μ^{-31} P K series photopeaks served as reference lines in the three prompt spectra. Hence, the dominant error when determining the nuclear charge radii differences was statistical. Regarding, however, the absolute energies, possible shifts in the positions of the calibration lines between the different time-gated spectra ("prompt," "delayed," and "calibration," with respect to a stopped muon) have to be corrected for. These shifts

amounted to 5–10 eV and could be measured with an accuracy of 6 eV. The nonlinearity of our measuring system was determined from well-known γ -ray transitions in radioactive ¹¹⁰Ag^m and ¹⁹²Ir sources^{11,12} in the interesting energy range from 300 to 700 keV. The same nonlinearity behavior was found in the calibration and the delayed spectra. In the latter, the calibration peaks fed through by accidental coincidences. Therefore, these nonlinearity corrections have been adopted for the prompt spectra.

As an example, Fig. 1 shows parts of the prompt spectra of ^{nat}S , ^{34}S , and ^{36}S taken with the 50 cm³ detector. The 511 keV annihiliation peak, although of considerable strength when little target material was available (³⁴S, ³⁶S), does not significantly enhance the error of the center-of-gravity positions of the respective 2p-1s photopeaks because its line shape can be accurately determined from the delayed spectrum. Table II shows the K series transition energies measured in the present work together with previous results of $\mu^{-31}P$ and $\mu^{-nat}S$ of our group.¹³ The listed energies are corrected for the abundance of the isotopes present in each target (see Table I) and for the recoil due to the emitted muonic x ray. The latter amounted to 4-7 eV. With the exception of the 4p-1s transition in ³¹P, there is good agreement between the different energies showing the consistency of the data. The 3p-1s transition in ³⁴S could not be reliably obtained, due to unidentified background. Since statistics in the 6.5 cm³ detector were generally poorer and, in particular, not sufficient to determine precise nonlinearity corrections, the final analysis has been done using the results of the 50

TABLE I. Isotopic compositions of the sulphur targets.

		Isotopic co	mposition (%)	
Target	³² S	³³ S	³⁴ S	³⁶ S
natS	95.02	0.75	4.21	0.02
³⁴ S	0.06	0.42	99.5	0.06
³⁶ S	0.13	0.01	18.8	81.1

			TAI	BLE II. Experim	ental energies of	the K-series trans	sitions.			
		³¹ P			³² S		34,	S	36	
	Present	t work		Present	work		Present	work	Present	work
Transition	50 cm^3	6 cm ³	Ref. 13	50 cm^3	6 cm ³	Ref. 13	50 cm^3	6 cm ³	50 cm^3	6 cm ³
2 <i>p</i> -1 <i>s</i>	456.806(12)	456.798(25)	456.831(20)	516.340(12)	516.315(23)	516.327(20)	516.110(16)	516.110(27)	515.985(14)	515.988(27)
3 <i>p</i> -1 <i>s</i>	544.866(13)	544.866(25)	544.912(20)	616.585(40)		616.578(30)			616.284(80)	
4 <i>p</i> -1 <i>s</i>	575.617(15)	575.691(40)	575.669(25)	651.610(35)		651.582(35)	651.358(140)		651.300(100)	
5p-1s	589.852(16)	589.887(40)	589.851(30)	667.832(35)		667.837(40)	667.577(110)		667.629(115)	
6 <i>p</i> -1 <i>s</i>	597.583(17)		597.601(30)							

 cm^3 diode. An independent analysis of the data of the 6.5 cm^3 Ge detector is, however, available as a diploma thesis.¹⁴ The quoted energy errors are a quadratic addition of statistical uncertainty, nonlinearity error, shift error between prompt and calibration spectra, and absolute uncertainties of the calibration lines. Regarding isotope shifts (see Table VI), absolute calibration and shift errors can be neglected.

In order to determine the nuclear charge radii and their differences, the measured transition energies have to be compared with calculations using the Dirac equation. Such calculations include all orders of electron-positron vacuum polarization, $\mu^+\mu^-$ and hadronic vacuum polarization, first and second-order vertex corrections, anomalous magnetic moment, relativistic reduced mass, electron screening, and nuclear polarization. The latter correction has been calculated by using the full sum rule strengths for the giant multipole resonances¹⁵ and the B(E2) excitation probabilities to the low lying rotational states.^{16,17} For ³¹P, the nuclear polarization correction amounts to 57 eV for the giant resonances and 9 eV for the low-lying states. For 32 S, the corresponding values are 70 and 17 eV; for ³⁴S, 71 and 10 eV; and for ³⁶S, 74 and 5 eV. The reason for the smaller corrections from the low lying states lies in the diminishing deformation at the end of the $1d_{3/2}$ neutron shell. The remaining difference between experimental and theoretical energies is due to the finite nuclear charge extension. In order to evaluate the nuclear charge radii from the finite size energy shift, we have employed the program MUON written by Rinker.¹⁵ This program solves numerically the Dirac equation for the muon nucleus system using a given nuclear charge density distribution. For convenience in the calculations, one generally employs the two-parameter Fermi distribution with a fixed skin thickness t=2.3 fm. The halfdensity radius c is varied by a least-squares fitting procedure until a minimum χ^2 value between experimental (E_{expt}) and theoretical (E_{th}) energies for the measured K-series transitions is obtained. The results in terms of the differences $E_{expt} - E_{th}$ and the relative experimental errors δE are shown in Table III. Also listed are the χ^2 values. For ${}^{31}P$, the 3p-1s and 4p-1s energies show a slight deviation between experiment and theory pointing to possibly unknown background. However, this is too small to influence the quoted errors. Table IV lists the results in terms of equivalent charge radii $R_{k,\alpha}$ and their sensibilities C_z , with respect to energy changes. The $R_{k,\alpha}$ radii first introduced by Barrett¹⁸ are model-independent quantities. This means that the form of the charge distribution used to calculate the $R_{k,\alpha}$ radius does not change its value within the experimental error.^{1,3} The k and α parameters also listed in Table IV have been determined from a least-squares fit to the muon generated potential.¹⁹ For the three sulphur isotopes, these parameters vary less than the last digit quoted in the table. The two errors for the equivalent radii $R_{k,\alpha}$ are the absolute errors of the experimentally measured transition energies and a quadratic addition of this uncertainty with a 30% error due to the nuclear polarization correction.

In order to evaluate model-independent rms radii, the ratio



FIG. 1. Parts of the prompt spectra of ^{nat}S, ³⁴S, and ³⁶S showing the respective muonic 2p-1s transitions, the $\mu^{-31}P$ reference lines, and the 511 keV annihilation peak.

	TABLE III. Least-	squares his to the x-se	ries transition energies.			
Contract of Content of	$(E_{\text{expt}} - E_{\text{th}}) \pm \delta E \text{ (eV)}$					
Transition	³¹ P	³² S	³⁴ S	³⁶ S		
2 <i>p</i> -1 <i>s</i>	-2 ± 7	-4±11	1± 15	-1 ± 12		
3 <i>p</i> -1 <i>s</i>	14± 8	15 ± 40		32± 80		
4p-1s	-14 ± 10	2±35	-43 ± 140	-4 ± 100		
5p-1s	-10 ± 11	26±35	-25 ± 110	121±115		
6p-1s	5±12					
<i>χ</i> ²	12.4	1.7	0.3	2.6		

TABLE III. Least-squares fits to the K-series transition energies

TABLE IV. Best fit parameters to the nuclear charge distribution.

TABLE V. Model-independent rms radii of the even sulphur isotopes and comparison with elastic electron scattering data.

Isotopes	k	α	$R_{k,\alpha}$ (fm)	C_z (am/eV)		<i>V</i> ₂	$\langle r^2 \rangle^{1/2}$ (fm)	
³¹ P	2.086	0.0522	4.0950(14:27)	-0.115	Isotope	Ref. 20	Present work	Ref. 5
³² S	2.093	0.0549	4.1875(11;26)	-0.089	³² S	1.284 05(9)	3.261(1)	3.248(11)
³⁴ S	2.093	0.0549	4.2172(14;26)	-0.089	³⁴ S	1.284 05(12)	3.284(2)	3.281(13)
³⁶ S	2.093	0.0549	4.2359(12;24)	-0.089	³⁶ S	1.284 66(9)	3.297(1)	3.278(11)

TABLE VI. Isotope shifts between ³²S, ³⁴S, and ³⁶S.

Isotope pair	1	Method	ΔE (eV)	$\Delta R_{k,\alpha}$ (am)	Δrms (am)
³⁴ S- ³² S	μ^{-} $\mu^{-} + (e,e)$ (e,e)	This work This work Refs. 5 and 21	-235±16	29.7±1.4	23.1 ± 1.2 32 ± 10
³⁶ S- ²⁴ S	$\mu^{-} + (e,e)$ (e,e)	This work This work Refs. 5 and 21	-123±17	18.7±1.5	13.0 ± 1.2 -7 ±12
³⁶ S- ³² S	$\mu^- + (e,e)$ (e,e)	This work This work Refs. 5 and 21	-358±13	48.4±1.2	36.1 ± 1.0 34 ± 10

TABLE VII. Comparison of Hartree-Fock calculations including ground state correlations with our experimental results.

			-	
	$\Delta \langle r$	$\binom{2}{0}^{1/2}$	$\Delta \langle r$	$(2)^{1/2}$
Force	³⁴ S- ³² S	³⁶ S- ³⁴ S	³⁴ S- ³² S	³⁶ S- ³⁴ S
Skyrme M	27 am	31 am	12±2 am	18±4 am
Skyrme G_{σ}	19 am	24 am	4±2 am	11±4 am
This work		•	23±1 am	13±1 am

$$V_2 = \frac{R_{k,\alpha}}{\mathrm{rms}}$$

from the recent elastic electron scattering results of Rychel *et al.*^{5,20} has been employed. Using this ratio, the muonic $R_{k,\alpha}$ radius can be converted model independently into the rms radius with almost no loss of accuracy. The V_2 ratios, together with the rms radii using our $R_{k,\alpha}$ values from Table IV, are listed for the three sulphur isotopes in Table V. The errors of these model-independent rms values correspond to a quadratic addition of $\delta R_{k,\alpha}$ and δV_2 . Finally, Table V contains the rms radii obtained only from electron scattering.⁵

Table VI shows the 2p-1s isotopic energy differences $\Delta E \pm \delta E$ calculated from the least-squares fits of Table III. These differences contain both the reduced mass shift (positive) and the field or actual isotope shift (negative). The last two columns list the isotope shifts between the three even sulphur isotopes, i.e., ³⁴S-³²S, ³⁶S-³⁴S, and ³⁶S-³²S, in terms of both equivalent charge radii differences $\Delta Rk, \alpha$ and model-independent root-mean-square radii differences Δrms . No nuclear polarization errors have been included, since for the differences these errors are small compared to the experimental errors. Also listed are the recent elastic electron scattering results of Rychel *et al.*⁵ They are in good agreement with the muonic data for the isotope pair ${}^{36-32}$ S, in fair agreement for ${}^{34-32}$ S, but significantly different for ${}^{36-34}$ S, even taking the much larger error of the (e,e) data into account. In particular, there is no decrease of the nuclear charge radius when adding the last two neutrons of the $1d_{3/2}$ shell.²¹

If we assume the nuclear extension to be simply proportional to $A^{1/3}$, the rms differences become considerably larger than measured. This fact has been known for quite some time. Angeli and Csatlos²² have given a refined semiempirical formula for the A dependence of the rms radii in which the nuclei are considered to be compressible. If we express this A dependence as in Ref. 23, i.e.,

$$\langle r^2 \rangle^{1/2} = r_0 A^{1/3} (1 + b A^{-2/3} - c A^{-4/3}),$$

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and employ Angeli's average values for the constants r_0 , b, and c, we obtain rms differences between the even S isotopes which are still a factor of 2 too large.

According to the approach of Reinhard and Drechsel,²⁴ a combination of spherical Hartree-Fock (HF) calculations with ground state correlations yields for the difference of the rms radii

$$\Delta \langle r^2 \rangle^{1/2} = \Delta \langle r_0^2 \rangle^{1/2} + \Delta \langle r_\beta^2 \rangle^{1/2}$$
$$= \Delta \langle r_0^2 \rangle^{1/2} + \frac{5}{8\pi} \langle r_0^2 \rangle^{1/2} \Delta \beta_2^2$$

where $\langle r_0^2 \rangle^{1/2} = \text{rms}$ radius from spherical HF calculations. The quadrupole deformation parameter β_2 is given by

$$\beta_2^2 = \left(\frac{4\pi}{5Z\langle r^2\rangle}\right)^2 B(E2) \ .$$

From the $B(E2,0^+\rightarrow 2^+)$ values for the first 2⁺ levels, namely $B(E2,^{32}S) = (304 \pm 11)$ fm⁴ (Ref. 16), $B(E2,^{34}S) = (194 \pm 8)$ fm⁴ (Ref. 25), and $B(E2,^{36}S) = (97\pm 26)$ fm⁴ (Ref. 16), we obtain $\Delta\beta_2^2(^{34}S^{-32}S) = -0.0233(20)$ and $\Delta\beta_2^2(^{36}S^{-34}S) = -0.0206(60)$. This means that the influence of the deformation alone yields a decrease in the difference of the rms radii of

$$\Delta \langle r_B^2 \rangle^{1/2} ({}^{34}\text{S} - {}^{32}\text{S}) = -15 \pm 2 \text{ am}$$

and

$$\Delta \langle r_B^2 \rangle^{1/2} ({}^{36}\text{S} - {}^{34}\text{S}) = -13 \pm 4 \text{ am}$$
.

Using two different Skyrme forces, Skyrme M and Skyrme G_{σ} (Ref. 26), one obtains the results given in Table VII. Although these forces are quite different²⁶—in particular, in G_{σ} ground state correlations are explicitly taken into account—both of them give an increase of 4–7 am for the radii differences

$$\Delta \langle r^2 \rangle^{1/2} ({}^{36}\text{S} - {}^{34}\text{S}) - \Delta \langle r^2 \rangle^{1/2} ({}^{34}\text{S} - {}^{32}\text{S})$$

independent of whether or not the quadrupolar deformation is taken into account. In contrast to those calculations, the experiment shows a decrease. Our experimental result, however, is in accordance with the systematic behavior of rms radii differences which show for a sequential addition of neutron pairs in the different shells, i.e., after N=20, 28, and 50, an almost linear decrease.²

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