Ultrasensitive Radioactive Detection of Collinear-Laser Optical Pumping: Measurement of the Nuclear Charge Radius of ^{50}Ca

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We report the first application of an ultrasensitive detection scheme in on-line collinear-laser spectroscopy. It is based on radioactive detection of optical depopulation pumping, using state-selective charge exchange as an intermediate step. This extends the calcium isotope shift measurements beyond the $f_{7/2}$ shell closure. The extracted ^{50}Ca mean-square charge radius constitutes the first experimentally determined one of a short-lived neutron-rich isotope beyond the $N=28$ shell closure. The steep increase in radius after the doubly magic ^{48}Ca nucleus indicates a strong coupling between the $p_{3/2}$ neutrons and the core protons.

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During the past decade, collinear-laser spectroscopy at on-line mass separators has provided a wealth of new information about nuclear spins, moments, and radii by the development and the application of techniques with increasing sensitivity. Recent methods exploit the strong optical depopulation pumping of the initial atomic (ionic) state to detect the optical resonances by state-selective charge-changing collisions and ion (atom) counting instead of the less efficient photon counting [1-5]. The sensitivity limit is in many cases determined by the presence of strong beams of stable or long-lived isobars, superimposed on the weak beams of short-lived isotopes. One type of solution to this background problem would be the development of element selective (laser) ion sources, or very-high-resolution mass separators. We used radioactive detection of the optically pumped ions after state-selective charge exchange (later referred to as ROC) to avoid the influence of stable isobaric contaminants. In this Letter, we report the first application of this method to extend the calcium isotope shift measurements beyond the $N=28$ shell closure, thereby also providing the first experimentally determined charge radius for a short-lived isotope in the $20 \leq Z \leq 28$, $N > 28$ mass region.

The charge distribution of the isotopes in the calcium chain has been investigated extensively (see Ref. [6] and references therein), so far covering all isotopes from ^{40}Ca to ^{48}Ca, both being doubly magic nuclei. All existing laser spectroscopy data were calibrated with muonic isotope shifts combined with electron scattering data in a comprehensive analysis [6], yielding a parabolic dependence of the mean-square charge radii as a function of neutron number, with a pronounced odd-even staggering. Several Hartree-Fock-based calculations have been performed by Caurier and Poves [7], Waroquier, Heyde, and Wenes [8], and Barranco and Broglia [9], finally resulting in a good agreement with experiment. Both the parabolic trend and the odd-even staggering were also reproduced by Talmi [10] using a shell-model-based theory.

An extension of the isotope shift and nuclear moment data across the $N=28$ shell closure is of fundamental interest as it provides a crucial test for Hartree-Fock calculations and for shell model descriptions far from stability. Nuclear spectroscopy experiments show, as expected in the shell model, a significant lowering of the energy of the first excited 2^+ nuclear state when going from the $N=28$ $_{20}$Ca, $_{22}$Ti, $_{24}$Cr, $_{26}$Fe, and $_{28}$Ni isotopes to the corresponding $N=30$ isotopes. The nuclear charge radii obtained from muonic isotope shifts for all stable isotopes in this region increase steadily above the $N=28$ shell closure [11,12]. For the closed-proton-shell calcium isotopes with $N > 28$, however, no charge radii could be measured so far due to the lack of sensitivity of the experimental methods used.

In this paper we report the isotope shifts in the Ca II transition $4s\,^2S_{1/2} \rightarrow 4p\,^2P_{1/2}$ at 397.0 nm and the deduced mean-square charge radii of eight calcium isotopes, including ^{50}Ca measured by ROC. This new method uses the first two steps of the previously reported optical pumping, state-selective charge exchange, particle counting scheme [1-5]. In the following we restrict the description to the specific case of our calcium measurements, and briefly sketch the straightforward generalization to other elements. After impact of the 600-MeV proton beam of the CERN synchrocyclotron on a hot uranium carbide target, neutron-rich calcium-ion beams were produced at the ISOLDE on-line isotope separator by a surface-ionization source, 34 kV acceleration and mass separation. The yield for ^{50}Ca amounted to 5×10^4 ions/s at a proton beam intensity of 2 μA. The ^{50}Ca beam was severely contaminated by isobaric beams with an intensity of the order of 10^8 ions/s, mainly consisting of stable titanium. The experimental setup is shown in Fig. 1. In the optical pumping zone of about 150 cm length, the selected ion beam is made to overlap with the

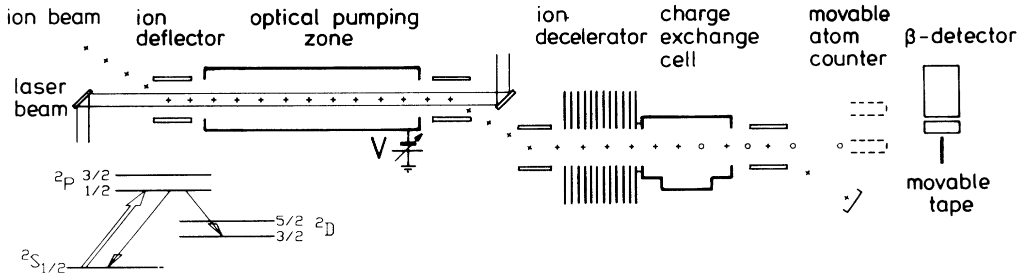

FIG. 1. Experimental setup. Inset: The low-energy electronic level scheme of Ca II and the laser optical pumping cycle are shown.

beam of a cw dye laser. The effective laser frequency is varied by changing the potential on the optical pumping zone. At resonance, repeated cycles of laser excitation $4s\,^2S_{1/2} \rightarrow 4p\,^2P_{1/2}$ and spontaneous decay, partially to the $4d\,^2D_{3/2}$ metastable state, finally leads to a strong depopulation of the ground state and a corresponding population of the metastable state (see inset of Fig. 1). This change in atomic state of the ions is transformed into a change in charge state by partial neutralization during passage through a sodium vapor. The ratio of the neutralization cross sections of the metastable and the ground state is strongly dependent on the ion impact energy and reaches a maximum value of 3 at about 4 keV [13]. Therefore the ions are decelerated to 4 keV prior to neutralization to exploit this velocity dependence. Finally, the remaining ions are deflected and the atoms are counted by detecting the secondary electrons emitted after impact on a movable metallic tape.

Although for pure calcium beams this detection scheme is extremely sensitive, the overwhelming stable background in the $A=50$ beam inhibits the measurement of the ^{50}Ca isotope shift by the atom counting technique. However, the nonradioactive contamination can be suppressed completely by detecting the β radiation of the decaying ^{50}Ca isotopes (lifetime 13.9 s) instead of the atoms themselves. A 2-mm plastic scintillator detector with a solid angle of 25% was used for this purpose. Every 20 s, a tape transport carried all radioactive nuclei away from the detectors, the effective laser frequency was changed, and a new counting period started. The reference spectra of ^{44}Ca and ^{46}Ca were taken immediately before and after the ^{50}Ca spectrum.

A spectrum obtained in a single 20-min scan is shown in Fig. 2. From the neutralization cross sections and the optical pumping efficiencies, a realistic estimate of the signal-to-background ratio (S/B) gives 100%. The reduction of this ratio to 40% is mainly due to the radioactive isobars in the reference beams spilling partially into the detection zone. This background also inhibited the ^{52}Ca measurement during the same measuring period. Shielding the β detector will yield a detection limit as low as 10 primary calcium ions/s (for a measuring time of 10 s/channel). For the odd calcium isotopes, the S/B of the $S_{1/2} \rightarrow P_{1/2}$ hyperfine components is reduced to 5% to 10%, which will still be above the required S/B for the presently available production rates of the isotopes ^{39}Ca and ^{49}Ca.

The ROC method with state-selective neutralization as the charge-exchange mechanism is also applicable to ion beams of short-lived isotopes of other alkaline-earth elements, as they all have similar low-lying ionic level schemes. It has an analogous and even more general applicability for laser spectroscopy on neutralized fast atom beams using state-selective reionization as the intermediate step [1]. It should be underlined that the present method exploits the optical depopulation pumping, which is in variance with the Zeeman pumping used in the experiments on radioactive detection of optical pumping performed on atomic vapors [14] and in the more recent measurements on fast atomic Li beams [15]. In the latter

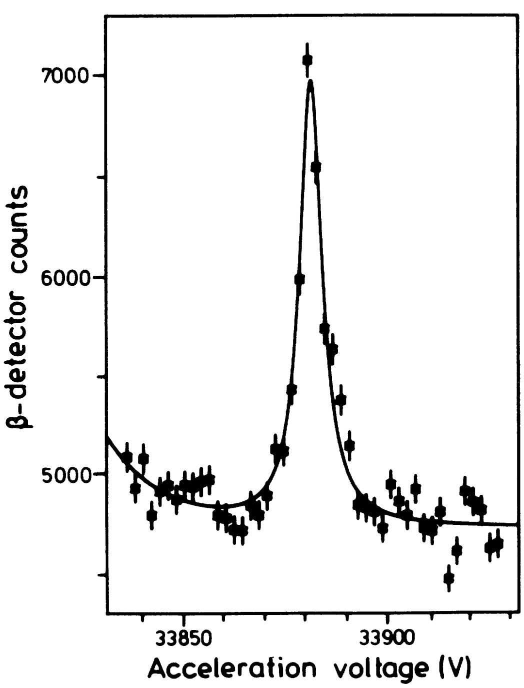

FIG. 2. Optical $4s\,^2S_{1/2} \rightarrow 4p\,^2P_{1/2}$ resonance signal of ^{50}Ca, measured by radioactive detection of metastable state population via optical pumping. The total measuring time was 20 min.

cases, the hyperfine interaction transfers the laser-induced atomic polarization or alignment into a nuclear orientation, which is monitored by the asymmetry of the emitted β or γ radiation. The ROC method, on the other hand, is based on an efficient atomic-state-to-charge-state conversion, and the optical pumping is monitored by the β count rate instead of the asymmetry.

The obtained isotope shift results are presented in Table I. A King plot combining the $\delta\langle r^2\rangle$ data from the analysis of Palmer *et al.* [6] with these isotope shift values provides a calibration [specific mass shift factor $K^{\mathrm{SMS}} = -9.2(3.8)$ GHzu, electronic field shift factor $F = -283(6)$ MHz/fm^2 [16]] to extract all $\delta\langle r^2\rangle^{44,A}$ from our data. Recent calculations [16] of this F factor perfectly agree with this evaluation. For consistency with earlier work the former procedure was used to extract the $\delta\langle r^2\rangle$ values; these are listed in Table I and plotted in Fig. 3.

As pointed out by Gräf *et al.* [17], there is a remarkable similarity in the neutron number dependence of the mean-square charge radii and the monopole matrix elements $0_1^+ \rightarrow 0_2^+$ which were both explained within the same model. On the other hand, Barranco and Broglia [9] calculated $\delta\langle r^2\rangle$ for the even-$f_{7/2}$-shell calcium nuclei in very good agreement with the experimental values by taking into account all collective deformation contributions from excitations to low-lying 0^+, 2^+, 4^+, 3^-, and 5^- levels. Wave functions for these states were calculated by Hartree-Fock (HF) methods and transition probabilities and dynamic deformations were extracted, from which changes in mean-square charge radii were calculated. This approach showed that especially for the nuclei close to ^{40}Ca, the odd-parity contributions to the mean-square charge radii are very important. Our measurement of the ^{50}Ca radius offers the possibility to extend the comparison of these HF-type calculations with experimental radii beyond the $N=28$ shell closure. Spherical HF+BCS calculations for the charge radii of Ca isotopes

TABLE I. Measured isotope shifts (IS) in the Ca II $4s\,^2S_{1/2} \rightarrow 4p\,^2P_{1/2}$ transition and derived changes in mean-square nuclear charge radii $\delta\langle r^2\rangle$. For the IS values the errors between parentheses are the random errors and those between square brackets include the systematic calibration error of 2 MHz per mass unit difference. The errors on the $\delta\langle r^2\rangle$ stem mainly from the random errors on the IS results.

A	IS^{44,A} (MHz)	$\delta\langle r^2\rangle^{44,A}$ (fm^2)
40	−842(3)[9]	−0.280(14)
42	−417(3)[5]	−0.078(11)
43	−170(8)[8]	−0.156(28)
44	0	0
45	249(3)[4]	−0.154(11)
46	445.2(0.6)[4]	−0.157(5)
48	854(3)[9]	−0.302(14)
50	1109(9)[15]	−0.007(34)

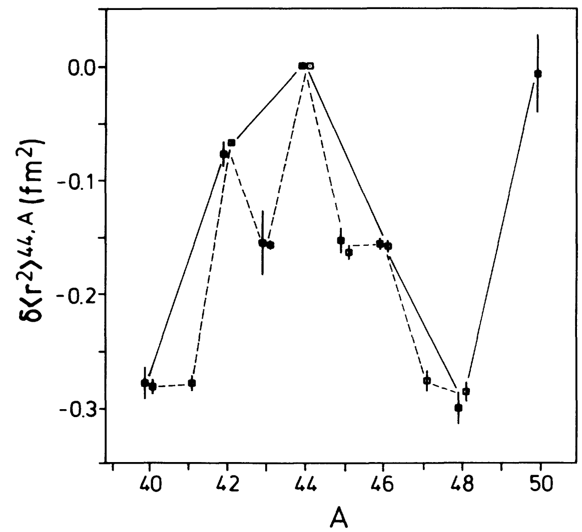

FIG. 3. Experimental changes in mean-square nuclear charge radii of the calcium isotopes. Solid squares represent the present data; open squares show the data from Ref. [6].

across the $N=20$ and 28 shell closures ($38 \leq A \leq 50$) have been carried out by Waroquier, Heyde, and Wenes [8]. As expected [8], the results are not able to describe the parabolic shape of the curve between $A=40$ and 48, but the shell effect at $N=28$ is well pronounced in those spherical calculations which account for roughly one-third of the observed $\delta\langle r^2\rangle$ between $N=28$ and 30.

The influence on the mean-square charge radius of the addition of a neutron pair is supposed to be smaller for the ^{48}Ca double magic nucleus than for isotones with valence protons above the $Z=20$ shell closure, in which a considerable interaction between added valence neutrons and the valence protons is expected. However, the obtained $\delta\langle r^2\rangle^{N=30,N=28}$ value for calcium turns out to be equal to those for chromium and iron [11,12]. This suggests that the increase in collectivity giving rise to the radius increase for the $N=30$ isotones is mainly due to a rearrangement of the ($Z=20$) core protons. A similar conclusion was drawn from the results of muonic measurements of radii for isotones with $N=24$, 26, and 28 [11].

In view of the similarity between the charge radii for all measured elements, the odd-even staggering effect for the sequences $^{48\text{-}49\text{-}50}$Ca and $^{52\text{-}53\text{-}54}$Cr are expected to be nearly the same. Andl *et al.* [18] quote a lower limit for the ^{49}Ca charge radius from the fact that no resonance attributable to ^{49}Ca was found in their experimental spectrum containing a very strong ^{48}Ca resonance. This value ($\delta\langle r^2\rangle^{49\text{-}48} \geq 0.5$ fm^2) would lead to an odd-even staggering for the calcium sequence which would be much stronger than for the Cr sequence and, in addition, would be inverted. A new measurement on ^{49}Ca is needed to get a reliable $\delta\langle r^2\rangle$.

The consistent behavior of the charge radii differences as a function of neutron number can be related to the known excitation energies of the first excited 2^+ nuclear states, which are much higher for the $N=28$ isotopes than for the neighboring isotopes, revealing a clear neutron shell closure effect for all elements studied. In this regard an interesting question arises concerning ^{52}Ca. The E_{2^+} of the iron and nickel isotopes with $N=32$ are roughly equal to the E_{2^+} of the corresponding $N=30$ isotopes, which is also reflected in the positive $\delta\langle r^2\rangle^{N=32,N=30}$. However, the first excited 2^+ state of ^{52}Ca lies 2.5 times higher [19] than the corresponding state in ^{50}Ca, suggesting a distinct $p_{3/2}$ subshell closure at ^{52}Ca. So a measurement of the mean-square charge radius of ^{52}Ca would be particularly interesting to clarify the semimagic character of this nucleus.

In conclusion, the ROC method allowed the extension of calcium nuclear charge radii measurements to ^{50}Ca, a nucleus outside the $f_{7/2}$ shell. The results show a pronounced shell effect at $N=28$ which is very similar in magnitude to the effect found for elements further away from the $Z=20$ shell closure. The applied new method creates important new perspectives for the laser spectroscopic study of short-lived nuclei very far from stability, which were up to now inaccessible by standard detection techniques.

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