Measuring the Variation in Nuclear Charge Radius of Xe Isotopes by EUV Spectroscopy of Highly-Charged Na-like Ions

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(Dated: June 26, 2018)

The variation in mean-square nuclear charge radius of xenon isotopes was measured utilizing a new method based on extreme ultraviolet spectroscopy of highly charged Na-like ions. The isotope shift of the Na-like D1 (3s 7S1/2 - 3p 2P1/2) transition between the 124Xe and 136Xe isotopes was experimentally determined using the electron beam ion trap facility at the National Institute of Standards and Technology. The mass shift and the field shift coefficients were calculated with enhanced precision by relativistic many-body perturbation theory and multi-configuration Dirac-Hartree-Fock method. The mean-square nuclear charge radius difference was found to be δ < r^2 >_{136Xe,124Xe} = 0.269(0.042) fm^2. Our result has smaller uncertainty than previous experimental results and agrees with the recommended value by Angeli and Marinova [I. Angeli and K. P. Marinova, At. Data and Nucl. Data Tables 99, 69-95 (2013)].

The charge radius and the mass of the nucleus are its most fundamental properties. Studies of nuclear charge radii essential to understanding nuclear structure have revealed unusual properties such as nuclear halos [1], contribute to precision tests of the Standard Model [2], and enter in the determination of stellar element abundances [3].

Only a few complementary techniques exist today for the determination of the absolute mean-square nuclear charge radius < r^2 >, and its isotope variation δ < r^2 >. Muonic-atom spectroscopy [4] has been highly successful in the absolute measurement of < r^2 >. Generally its accuracy is limited by large nuclear polarization effects as the muon orbit is comparable to the nuclear size. Electron scattering has also been widely used for the determination of the same quantity in heavy nuclei [5] where the challenge is that the experimental cross-sections have to be analyzed beyond the first Born approximation to take into account the phase shift. Both methods require considerable amounts of target material and with exception of recent efforts [6], are generally applied to stable nuclei.

X-ray spectroscopy of inner-shell Kα lines and valence-electron optical isotope shifts allow for δ < r^2 > measurements between isotopes [7]. The laser spectroscopy measurements of the latter technique offer utmost experimental precision and can be applied to long chains of stable and unstable isotopes. The difficulty of this technique lies in the calculation of the electronic structure of many-electron atomic systems that often include electron correlation effects and can contribute to systematic offsets in the inferred δ < r^2 > [8]. Electron screening or correlation effects in heavy elements, such as bismuth or uranium, can be particularly difficult to calculate theoretically. These calculations are sometimes benchmarked by non-optical methods such as Kα measurements [9] or King plot analyses [10].

In the search for new methods for the measurement of nuclear radii, particular charge states of highly ionized atoms have been considered due to their simpler electronic structure and higher sensitivity to the nuclear charge distribution, because the compressed electron cloud can produce large isotope shifts of energy levels. Relativistic normal and specific mass shifts were explored through magnetic-dipole transitions of Be-like and B-like argon isotopes in the visible range [11], but the experimental precision was insufficient to probe the charge distribution. Precision X-ray spectroscopy [12] and dielecrtronic recombination measurements [13, 14] of isotope shifts in heavy, few-electron ions, have been used to determine δ < r^2 > in a variety of nuclei because the electronic structure of these ions can be calculated with high accuracy. However, the systems studied are generally difficult to produce experimentally, in particular for radioactive isotopes.

In this letter we introduce a new method based on accurate theoretical calculations for low-lying energy levels of Na-like ions. The simple 3s electronic configuration in these systems penetrates the Ne-like closed shell to probe the nucleus. Spectroscopic measurements of extreme ultraviolet (EUV) transitions are sensitive enough to determine nuclear charge distribution as previously discussed by Gillaspy et al. [15]. Atomic-structure calculations for these systems can reach accuracies higher than those for neutral atoms and singly charged ions used in optical isotope shift measurements.
Here, we present the first experiment using this technique to determine $\delta < r^2 >^{A,A'}$ of xenon isotopes by combining accurate theoretical calculations with precise measurement of the isotope shift in the frequency of the $3s\ 2S_{1/2} - 3p\ 2P_{1/2}$ (D1) transition in highly charged Na-like $^{136}\text{Xe}^{43+}$ and $^{124}\text{Xe}^{43+}$ ions. Benchmarks of the quantity for this isotope pair are the previous optical isotope shift measurement that reported a value of $0.242(0.008)$ fm$^2$ [10] and the recommended value of $0.2897(0.0050)$ fm$^2$ that considers interconnected trends across the nuclear radii surface in a compilation by [17]. The new technique uses an electron-beam ion trap (EBIT) that can be also employed for unstable nuclei as demonstrated by Elliott et al. [12].

The quantity determined experimentally is the $\delta\nu$ isotope-dependent frequency shift, which has two components:

$$\delta\nu = \delta\nu_{MS} + \delta\nu_{FS}. \tag{1}$$

$\delta\nu_{MS}$ is the mass shift due to the finite mass of the nucleus and $\delta\nu_{FS}$ is the field shift associated with the nuclear volume. It is notable that the field shift scales with the nuclear charge as approximately $Z^{8/3}$ and it dominates the mass shift in heavy systems. As an approximation, $\delta\nu_{FS}$ can be considered to be proportional to the difference between the mean-square nuclear charge radii of the two isotopes:

$$\delta\nu_{FS} = F\delta < r^2 >, \tag{2}$$

where $\delta < r^2 >$ is defined as [17]:

$$\delta < r^2 >^{A,A'} = < r^2 >^A - < r^2 >^{A'}. \tag{3}$$

Both the $F$ field shift coefficient and the $\delta\nu_{MS}$ mass shift can be obtained from highly accurate atomic-structure calculations allowing for the experimental determination of $\delta < r^2 >^{A,A'}$ from the measured $\delta\nu$ shift.

In this experiment, EUV spectra were collected for Na-like $^{136}\text{Xe}^{43+}$ and $^{124}\text{Xe}^{43+}$ ions produced in the EBIT at the National Institute of Standards and Technology (NIST). Details of the measurements of EUV emission from xenon ions are similar to that in previous experiments in this wavelength region [15].

Briefly, over the course of the experimental campaign, isotopically pure $^{136}\text{Xe}$ and $^{124}\text{Xe}$ neutral gases were alternately injected into the EBIT for approximately one-hour periods at a time. For each gas injection, a series of spectra were collected for five minutes each, using a liquid-nitrogen-cooled EUV charge-coupled-device (CCD) camera attached to a flat-field EUV grating spectrometer [18, 19].

The EBIT was operated at 6.0 keV electron beam energy and 150 mA electron beam current to optimize the production of the Na-like Xe charge state. The Na-like D1 transition was selected for the determination of $\delta < r^2 >^{A,A'}$ because of the high accuracy of the calculations for this line and because it is cleanly separated from other features in the EUV spectrum. The other major Na-like candidate, the $3s\ 2S_{1/2} - 3p\ 2P_{3/2}$ (D2) line, was found to be effected by a blend [20]. Figure 1 shows the full spectral range detected by the CCD camera, including emission from Na-like Xe and nearby charge states.

Absolute wavelength calibration was carried out as a regular procedure [19] using well-known transitions in different charge states of Ne, Xe, and Ar [21] collected several times during the data-taking epoch. The first derivative of the absolute calibration function provided the dispersion function to convert the measured spatial shift on the CCD chip to a wavelength shift. Measurement of the isotope shift of the D1 transition in this experimental arrangement entails the determination of a displacement that is well within the uncertainty of the absolute wavelength value of 12.3935(0.0009) nm.

Because of the less than $10^{-4}$ nm anticipated shift between the D1 lines of the two xenon isotopes, a large number of EUV photons were required to achieve the necessary statistical uncertainty. Owing to the count rate of the D1 line emission, the experiment was a multi-day acquisition effort during which long-term thermal and electronic systematic drifts could be expected. We adopted a data analysis procedure that allowed us to eliminate the long-term variation of the D1 line position by determining the centroid positions in channels (a column of CCD pixels) in each individual 300 s spectrum and creating a time-ordered sequence of the data that included both isotopes.

The full sequence of 428 values was fitted to a set of third order polynomial functions that described the long-term variation of the absolute position of the spectral line. Different sets of coefficients were allowed for data sets in between liquid nitrogen refills of the CCD camera at which times more thermal variations were expected. The polynomial functions for the two isotopes were kept to be the same except for an overall constant

![FIG. 1. Sample of a five-minute EUV spectrum of $^{136}\text{Xe}$ with spectral lines from different charge states. The inset shows the blowup of the Na-like D1 transition in first order of diffraction with a Gaussian fit.](image-url)
free parameter representing the isotope shift.

Figure 2a shows a partial series of the centroid values with the alternating isotope sequences, the polynomial fit, and the constant isotope shift. Residuals were binned for each isotope individually, providing statistical distributions, which were fit with pure Gaussian functions shown in Fig. 2b. The agreement between the centroids of the Gaussian functions is more than order of magnitude better than the uncertainty of the shift, giving us confidence in the evaluation procedure.

The \( \delta \nu_{MS} \) mass shift (MS) and \( \delta \nu_{FS} \) field shift (FS) for the Na-like D1 \((3s\,^2S_{1/2} - 3p\,^2P_{1/2})\) transition were calculated using two different theoretical methods: the relativistic many-body perturbation theory (RMBPT) \cite{22, 23} and the multi-configuration Dirac-Hartree-Fock (MCDHF) theory of the GRASP2K code \cite{24}.

RMBPT was performed up to third order including both the Coulomb and Breit interactions in each order. A relativistic configuration interaction (RCI) module was used in the GRASP2K code to consider the Breit interaction perturbatively as well as leading quantum electrodynamics (QED) contributions (vacuum polarization as well as self-energy corrections). It was found that the two theories were in an overall excellent agreement. Table II shows the experimental and calculated isotope shifts from the current investigation along with the calculations from the large-scale configuration-interaction Dirac-Fock (CIDF) method of Tupitsyn et al., which solves the Dirac-Coulomb-Breit equation to calculate the energies and isotope shifts \cite{25}.

To obtain the field shift from the experimental \( \delta \nu \) frequency shift, the mass shift was accounted for through theoretical calculations. In RMBPT theory, the relativistic nuclear recoil corrections were calculated up to order \((Z\alpha)^3\) beyond the nonrelativistic mass shift by using the Palmer operator \cite{26}. This operator gives the one- and two-body nuclear-recoil terms in the relativistic Hamiltonian corresponding to the \( \delta \nu_{NMS} \) normal mass shift (NMS) and \( \delta \nu_{SMS} \) specific mass shift (SMS), respectively. The nonrelativistic nuclear-recoil effect is itself of the order of \((Z\alpha)^3\), therefore, the leading relativistic correction considered here is of the order of \((Z\alpha)^4\). The mass shift in each order of RMBPT was found by taking the difference of two calculations, one including nuclear recoil throughout, and the other excluding nuclear recoil, and then carefully testing the difference for numerical significance. In this way, it was possible to infer the NMS and the SMS coefficients, \( R \) and \( S \), respectively, which are defined such that the \( \delta \nu_{NMS} \) and \( \delta \nu_{SMS} \) frequency shifts for two different isotopes with nuclear masses \( M_A \) and \( M_A' \) are given by:

\begin{equation}
\delta \nu_{NMS}(A, A') = R(1/M_A - 1/M_{A'}),
\end{equation}

and

\begin{equation}
\delta \nu_{SMS}(A, A') = S(1/M_A - 1/M_{A'}),
\end{equation}

where

\begin{equation}
\delta \nu_{MS} = \delta \nu_{NMS} + \delta \nu_{SMS}.
\end{equation}

Third-order RMBPT contributions to the mass shift coefficients for the D1 transition were found to be on the order 0.1 % or less of the total mass shift. The dominant theoretical uncertainty in the mass shift is likely to be associated with the omitted higher-order relativistic...
terms, which start at the order of \((Z\alpha)^4\) \cite{27}, and could be on the order of several percent of the total mass shift for the D1 transition. We assume a relative theoretical uncertainty of 5% throughout the calculations.

In order to calculate the field shift coefficient \(F\) in RMBPT, the transition frequency \(\delta\nu\) was calculated assuming different nuclear charge distributions for isotopes \(A = 136\) and \(A' = 124\) and the difference \(\delta\nu_{FS} = \delta\nu_{136} - \delta\nu_{124}\) was taken in each order. For the present calculations a two-parameter (\(c\) and \(a\)) Fermi model was assumed to obtain the nuclear potential experienced by the electrons. Parameter \(c\) is the half density radius \cite{28} and \(a\) is related to the surface thickness \(t\) of the charge distribution through the relation \(a = t/4ln3\). The RMBPT calculations for the field shift were found to converge rapidly, with the third-order contributions to the D1 transition accounting for less than 0.1% of the total \(\delta\nu_{FS}\). The field-shift coefficient is obtained as \(F = \delta\nu_{FS}/\delta < r^2 >\), where \(\delta < r^2 >\) is the change in mean-square radius of the two nuclear charge distributions assumed in the calculation.

The dominant theoretical uncertainty in the calculation of \(F\) comes from the unknown nuclear charge distribution. To estimate this nuclear-model dependence, the whole calculation procedure was repeated for several pairs of nuclear charge distributions, including changes of the charge density both on the surface and in the bulk volume regions of the nucleus. Through these changes, \(F\) fluctuated at the 2% level, which was assumed as the associated uncertainty.

In the MCDHF approach, the atomic state function was expanded in the configuration state functions of the same parity, total angular momentum \((J)\), and its projection \((M_J)\). The reference configurations were 1s\(^2\)2s\(^2\)2p\(^6\)3s and 1s\(^2\)2s\(^2\)2p\(^6\)3p for the ground state and excited state, respectively, and the single configuration Dirac-Fock state functions were calculated for the \(^{136}\)Xe isotope. The 1s, 2s, and 2p orbitals were treated as core while the 3s and 3p orbitals were treated as valence orbitals. Excitations from the 1s orbital were limited to single excitations to the active sets. The configuration state function space was expanded by means of single, double excitations from the occupied orbitals to active sets of up to \(n = 6\) and also triple excitations within the \(n = 3\) states to consider the valence-valence, core-valence, and core-core correlation effects. Virtual orbitals were added layer by layer to monitor the convergence of energies and oscillator strengths. The Breit interactions and the leading QED effects up to \(n = 5\) were also included during RCI calculations. Self-energy and vacuum polarization QED corrections were estimated phenomenologically and were found to enter at the 0.1% level.

The relativistic isotope shift (RIS3) module \cite{29} was used to calculate the mass shift from the wave functions. Similar to RMBPT, GRASP2K also includes nuclear recoil corrections of the order of \((Z\alpha)^4\) for mass shift calculations. The field shift was calculated explicitly from the difference between transition energies that are obtained by solving the MCDHF and Breit equations for isotopes \(A\) and \(A'\) separately, keeping the nuclear surface thickness the same (2.30 fm) for both.

For a transition involving a valence s electron, Eq. \(2\) can be more accurately replaced by

\[
\delta\nu_{FS} = F\lambda, \tag{7}\]

where \(\lambda\) is the Seltzer moment of the nucleus \cite{30}:

\[
\lambda = \delta < r^2 > + S_4 \delta < r^4 > + S_6 \delta < r^6 > = [1 + S_{HO}/\delta < r^2 >] \delta < r^2 >, \]

with \(S_{HO} = S_4 \delta < r^4 > + S_6 \delta < r^6 >\) representing the higher nuclear moment terms. This allows evaluation of the effect of higher order nuclear moments, and the values of the \(S_4\) and \(S_6\) coefficients \cite{30} suggest that these contributions to \(\delta\nu_{FS}\) is about 4% in our case.

Similar conclusions can be drawn from the procedure described by Li et al. \cite{31}, which uses the probability density of the electron wave function from the GRASP2K code at the origin to calculate the field shift, effectively selecting the term \(\delta < r^2 >\) in \(\lambda\). The field shift obtained this way was found to be 4% larger than the result implicitly containing higher-order nuclear moments described above.

| Theory       | RMBPT | GRASP2K CIDF [25] | Experiment |
|--------------|-------|-------------------|------------|
| Coefficients | \(\delta\lambda\) | \(\Delta\delta\lambda\) | \(\delta\lambda\) | \(\Delta\delta\lambda\) | \(\delta\lambda\) | \(\Delta\delta\lambda\) |
| NMS          | -4.8  | -4.8              | -4.8       | -4.8       | -4.8 | -4.8 |
| SMS          | -62.2 | -62.3             | -62.3      | -62.7      | -62.7 |
| Total MS     | -67.0 | -67.1             | -67.5      | 143.0      | 143.0 |
| FS           | 143.0 | 142.0             | 143.0      | 76.1       | 75.3 |
| Total        | 76.1  | 4.4               | 75.3       | 4.4        | 75.8 | 65.5 |

Using the calculated values of the mass shift and \(F\), along with the experimentally obtained frequency shift \(\delta\nu\), the difference between the mean-square nuclear charge radii of \(^{136}\)Xe and \(^{124}\)Xe was determined. The value obtained using the RMBPT theoretical method was 0.268(0.042) fm\(^2\) and using the GRASP2K results it was 0.270(0.042) fm\(^2\). Our reported value here is their average of \(\delta < r^2 >_{^{136}\text{Xe} - ^{124}\text{Xe}} = 0.269(0.042)\) fm\(^2\).

The overall one sigma reported uncertainty \(\Delta \delta < r^2 >\) includes uncertainties from the experimental shift, the mass shift correction, the field shift calculation, and the higher order nuclear moments:
The 16 % relative total uncertainty of the current determination of $\delta < r^2$ between $^{136}$Xe and $^{124}$Xe isotopes is dominated by the experimental uncertainty over the theoretical uncertainty, which amounts to about 3 % including the mass-shift. Details of the different quantities that contribute to the evaluation of $\delta < r^2$ together with their measured or estimated uncertainties are listed in Table I.

Figure 3 presents the result of the current experiment compared to five other previous values using various techniques. Our result agrees within its uncertainty with the recommended values of $0.2897(0.0050)$ fm$^2$ [17] obtained by detailed analysis of published experimental nuclear charge-radius data. It also agrees within the uncertainties with $\delta < r^2 >^{136,124} = 0.242(0.080)$ fm$^2$ obtained from the most recent optical (laser spectroscopy) isotope shift measurement by Borchers et al. [16]. Their combined 0.080 fm$^2$ uncertainty is the quadrature sum of the experimental uncertainty of 0.005 fm$^2$ and theoretical uncertainty of 0.080 fm$^2$. The latter is an order of magnitude larger than the reported experimental uncertainty, due to the lack of precise theoretical calculations for the neutral system. Our result is slightly outside the value of $0.350(0.030)$ fm$^2$ [8] obtained through a King plot analysis of optical isotope shifts [16] versus muonic-atom spectroscopy measurements [4].

FIG. 3. $\delta < r^2 >^{136}$Xe and $^{124}$Xe measured in this work through Na-like D1 EUV spectroscopy (open diamond) compared with previous measurements and analyses (circles). Rec.: recommended value by [17] (open circle); Muon: muonic-atom spectroscopy [4]; Optical-O1: optical shift by laser-spectroscopy [16]; King (O1-Mu): King plot analysis combining the optical measurements with the muonic-atom results [8]; O2-HF and O2-FS: optical (interferometer) shift based on the Hartree-Fock method and Fermi-Segré calculations [32]. The two horizontal lines represent the uncertainty listed for the recommended value [17].

In conclusion, the agreement of our measurement with the recommended value and previous measurements, shows that EUV spectroscopy of Na-like ions is a viable method, and has the potential to provide accurate nuclear-size measurements for heavy nuclei. Na-like ions can be produced in large abundance in small laboratory scale devices of only a million ions or less, offering the possibility of conducting isotope shift measurements on radioactive isotopes at existing rare-isotope beam facilities like the Isotope Separator and Accelerator (ISAC) facility at TRIUMF, and next-generation facilities such as the Facility for Rare Isotope Beams (FRIB) at Michigan State University.

This work was partially funded by the NIST Grant Award # 70NANB16H204 of the Measurement Science and Engineering (MSE) Research Grant Programs. AL and ACCV acknowledges support by the National Science Foundation under Grant No. PHY-1565546. JMD acknowledges funding from the National Research Council Research Associateship Award at NIST. GG acknowledges support by NSERC (Canada). ABJr acknowledges support of the US Department of Commerce and NIST under the program # G-3-0034. We would like to thank David Takacs for his help with the initial data analysis.

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