Measurements of the differences in the nuclear charge radii among uranium isotopes

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(Received 25 September 1997)

Based on precise spectroscopic measurements of the x-ray transitions of few-electron uranium ions, the difference in the nuclear charge radii of ^{235}U and ^{238}U has been determined. The measurement makes use of the simplified atomic structure of highly charged ions, and it is shown that the spectroscopic data from few-electron ions are readily interpreted in terms of the variation in the mean nuclear radius. A value of $\delta\langle r^2\rangle^{235,238} = -0.250 \pm 0.032$ fm^2 is found. Combining the results with data from earlier optical and x-ray measurements, values for $\delta\langle r^2\rangle^{A,238}$ with $A = 233$, 234, 235, and 236 are derived that are of higher accuracy than previous values. [S0556-2813(98)02802-7]

PACS number(s): 21.10.Ft, 27.90.+b, 32.30.Rj, 29.25.Ni

INTRODUCTION

Measurements of the nuclear charge radius provide important data on the collective structure of the nucleus. Electron scattering has provided a value for the root mean square charge radius $\langle r^2\rangle^{1/2}$, but only for ^{238}U [1]. Data for other isotopes have been provided by studying the differences $\delta\langle r^2\rangle$ of the nuclear charge radii among different isotopes relative to that of ^{238}U, where $\delta\langle r^2\rangle$ is defined in terms of the root mean square radius $r_{\rm rms}$ as $(r^2_{\rm rms})^A - (r^2_{\rm rms})^{238}$.

In earlier studies, the parameter $\delta\langle r^2\rangle$ has been inferred in the high-Z region from neutral-atom optical isotope shift studies [2] and muonic-atom x rays [3]. Neutral-atom optical and $K\alpha$ x-ray transition isotope shifts are sensitive to the same nuclear parameters. However, the $K\alpha$ results are easier to interpret in terms of $\delta\langle r^2\rangle$. There is a large theoretical uncertainty in the optical-transition specific mass shift which makes it difficult to deduce nuclear parameters from those measurements. For example, the best uranium $\delta\langle r^2\rangle^{233,238}$ value results come from Ref. [4]. Because the configuration interaction is difficult to calculate, they used $K\alpha$ data [5] to calibrate the optical line shift $\Delta E^{233,238}$ in terms of $\delta\langle r^2\rangle^{233,238}$. Thus, even though laser spectroscopy gave a very precise value of ΔE, the uncertainty in $\delta\langle r^2\rangle$ is determined by the less precise $K\alpha$ data. As a result, the four optical $\delta\langle r^2\rangle^{A,238}$ measurements ($A = 233$, 234, 235, and 236) are dependent on the lone value of $\delta\langle r^2\rangle^{233,238}$ from Ref. [5]. Since the $K\alpha$ measurements have difficult corrections of their own, including, for example, complications due to satellite transitions, this relationship has the potential of producing erroneous results.

In muonic atoms the muon wave function extends much deeper into the nucleus than would that of a bound electron. Therefore higher moments in the nuclear charge density distribution ($\delta\langle r^4\rangle$, $\delta\langle r^6\rangle$, etc.) affect the shift in transition energy, ΔE. In contrast, isotope shifts involving electronic transitions are primarily sensitive to the first moment $\delta\langle r^2\rangle$. Furthermore, vacuum polarization and nuclear polarization effects contribute a much larger share to muonic energy levels than for electronic levels. Thus, while muonic-atom measurements have provided data on a number of high-Z isotopes, they are complimentary to the atomic-transition data [6]. Table I summarizes the experimental data within the U system.

In a recent paper [7], we demonstrated the utility of trapping few-electron, very-high-Z radioactive ions in an electron beam ion trap (EBIT) for measuring the changes of the nuclear charge radii between different isotopes of the same element. Using precision x-ray spectroscopy and exploiting the simplified electronic structure of few-electron ions, Ref. [7] isolated the nuclear effects and inferred the isotopic variation of the nuclear charge distribution $\delta\langle r^2\rangle$ between ^{233}U and ^{238}U. In the present paper, we use this new, technique to measure $\delta\langle r^2\rangle$ in the ^{235}U and ^{238}U pair. We also revisited our measurement of the ^{235}U and ^{238}U pair [8] in light of revised theoretical values for the nuclear polarization [9]. Using our measurement of $\delta\langle r^2\rangle^{235,238}$ as well as our measurement of $\delta\langle r^2\rangle^{233,238}$, we reexamine earlier optical measurements and derive values of $\delta\langle r^2\rangle$ for several additional uranium isotopes.

TABLE I. Measured values of $\delta\langle r^2\rangle^{A,238}$. The $K\alpha$/optical values come from Refs. [4,5], and the muonic-atom values come from Ref. [6]. The EBIT value for $\delta\langle r^2\rangle^{235,238}$ represents present work; the EBIT value for $\delta\langle r^2\rangle^{233,238}$ is from Refs. [7,8]. The quoted uncertainties are 68% confidence limits.

A	Muonic atom (fm^2)	$K\alpha$/optical (fm^2)	EBIT (fm^2)
233	-0.520 ± 0.081	-0.433 ± 0.050	-0.432 ± 0.043
234	-0.368 ± 0.045	-0.331 ± 0.038	
235	-0.305 ± 0.042	-0.278 ± 0.032	-0.250 ± 0.032
236		-0.166 ± 0.019	

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TABLE II. The ^{238}U energy values and the nomenclature for the key are from Ref. [13]. The uncertainties represent 90% confidence limits, as discussed in Ref. [15]. All transitions decay to the ground state of the respective ion.

Key	Ion	Upper level	^{238}U energy (eV)
Li	U^{89+}	$(2p_{3/2})_{j=3/2}$	4459.37±0.35
Be	U^{88+}	$(2s_{1/2}2p_{1/2})_{j=1}$	4501.72±0.27
B-1,2	U^{87+}	$(2s_{1/2}2p_{1/2}2p_{3/2})_{j=1/2,3/2}$ blend	4521.39±0.22
C	U^{86+}	$(2s_{1/2}2p_{1/2}^2 2p_{3/2})_{j=1}$	4548.32±0.20
O-1	U^{84+}	$(2s_{1/2}2p_{1/2}^2 2p_{3/2}^3)_{j=2}$	4525.26±0.25

A reexamination of the variation of the nuclear charge radii in U is very timely in light of recent atomic-structure measurements that have focused on this element, i.e., the heaviest naturally occurring element. It was noted [10] that estimates for the radii of high-Z nuclei [11] were low when compared to values from muonic-atom studies. This issue is of critical importance in studies of quantum electrodynamic (QED) corrections to atomic energy levels in the high field of few-electron, high-Z ions. In atoms of $Z>90$, the nuclear size correction to the Coulomb energy is comparable to that of the QED contributions and thus is a large fraction of the Lamb shift. As a result, the uncertainty in the nuclear radius has dominated the uncertainty in theoretical estimates of the Lamb shift. This situation led to the conclusion that ''it would be worthwhile to measure the Lamb shift for various isotopes of high-Z elements in order to disentangle the nuclear size effects from the radiative corrections'' [11]. Furthermore, only recently have nuclear polarization calculations been done for few-electron, high-Z atoms for various isotopes [9,12]. These calculations have a large uncertainty due to incomplete knowledge of the input nuclear parameters. Although experimentally measurements of $2s$-$2p$ transitions in few-electron ions [13,14] are nearing the precision necessary to check these calculations, the results of Ref. [9] indicate it will be more difficult than previously thought to measure the nuclear polarization contribution. Accurate measurements of nuclear parameters are thus crucial not only for determining the fundamental properties of nuclear structure, but also for interpreting the measurements of atomic structure and of quantum electrodynamical effects.

EXPERIMENT

Our technique for determining $\delta\langle r^2\rangle$ is based on precise Doppler-shift-free measurements of the $n=2$ to $n=2$ x-ray transitions in nearly bare ions of the isotopes in question. Implementation of this technique has been made possible by the advent of a facility at which the x-ray transitions from such highly stripped, often radioactive, ions could be generated and measured conveniently and reliably. The transitions studied are the electric dipole, $2s_{1/2}$-$2p_{3/2}$ transitions in the three-electron Li-like ion, the four-electron Be-like ion, the five-electron B-like ion, and the six-electron C-like ion. Table II lists the specific transitions studied and their previously measured energies in ^{238}U [13,15]. Because the measurements are for transitions in an inner shell, the electron wave function overlap, especially that of the $2s$ electron, with the nucleus is large. It is thus an excellent probe of the nuclear charge distribution resulting in a relatively large energy shift (ΔE) as different isotopes are measured. Compared to muonic atoms, however, the overlap is modest, and large nuclear polarization corrections are avoided. Moreover, the atomic physics of few-electron ions is tractable and deducing $\delta\langle r^2\rangle$ from ΔE is relatively simple. Most importantly, it is not complicated by large specific mass shift corrections necessary in neutral ions [16]. In other words, in our measurement the coulomb shift (δE_{Coul}), which is directly related to $\delta\langle r^2\rangle$, is by far the dominant contribution to ΔE, and other atomic or nuclear contributions are minimal ($\leqslant 1\%$). A further benefit of our technique is that the energy of the $\Delta n=0$ transitions studied falls within a range where high-precision crystal spectroscopy is easily employed.

The measurements were done at the high-energy electron beam ion trap (SuperEBIT) at Lawrence Livermore National Laboratory [17]. An electron beam ionizes, excites, and radially traps the ions. The ions are trapped axially by potential differences between three colinear cylindrical electrodes through which the beam passes. Atoms or low-charge ions, injected into the trap, are ionized to high-charge states by successive collisions with beam electrons.

Like the ^{233}U ions studied in Ref. [7], the ^{235}U ions were provided by a novel source developed at LLNL [18] consisting of a thin wire platinum probe with a plated tip placed near the electron beam. It provided a continuous source of ions for the trap. The total mass of plated ^{235}U was 20 mgm. The ^{235}U had been isotopically enriched to 99.77%. This probe technique permits the study of any isotope which can be handled and plated in nanogram or greater quantities. The ^{238}U ions were provided by a metal vapor vacuum arc (MEVVA) source [19] using a ^{238}U cathode depleted in ^{235}U weighing 14 g.

The ions were studied by their characteristic x rays observed through ports in the cryogenic vessels surrounding the trap. The $2s_{1/2}$-$2p_{3/2}$ electric dipole transitions, situated near 4.5 keV, were analyzed in a high-resolution von Hámos-type curved-crystal spectrometer [20]. The spectrometer used a $120\times 50\times 0.25$ mm^3 LiF(200) crystal ($2d=4.027$ Å) bent to a 75-cm radius of curvature. X rays were recorded with a gas-filled position sensitive proportional counter with a $10\times 3\times 1$ cm^3 active volume. The working gas consists of 90% Xe with the balance isobutane at 15 psi overpressure. For the present measurements the nominal Bragg angle was 43.5°. The energy resolution of the setup was 1.1 eV full width at half maximum (FWHM), i.e., identical to that used in Ref. [7] in the measurement of ^{233}U.

Similar to the experimental procedure in Ref. [7], the x-ray spectrum of ^{235}U was compared with that from ^{238}U. Figure 1 shows the measured spectra of ^{235}U and ^{238}U. For comparison, we also included the spectrum of ^{233}U measured earlier in the figure. Each spectrum shown was accumulated over a total of approximately 150 h at a beam energy of 135 keV and current of 180–220 mA. The charge state distribution was somewhat more peaked about the Be-like ionization state in the ^{233,235}U data than for the ^{238}U data, which peaked about the B-like ionization state. The ^{238}U spectrum also shows a weak O-like line which is absent in the ^{233,235}U data. The difference in the ionization balance is the result of somewhat different trapping parameters used when injecting with

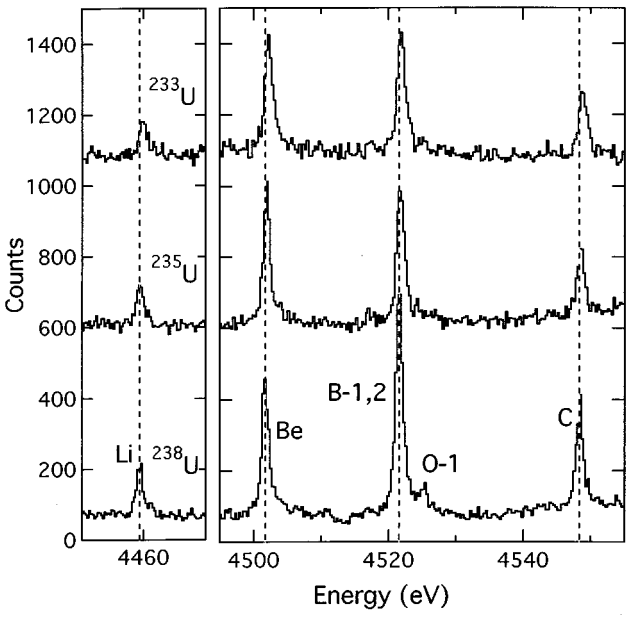

FIG. 1. Crystal spectrometer spectra of the $2s_{1/2}$-$2p_{3/2}$ transitions in U^{86+} through U^{89+} for the three isotopes ^{233}U, ^{235}U, and ^{238}U. The lines are labeled by the charge state of the ion; i.e., Li indicates a transition in Li-like U^{89+}, Be indicates a transition in Be-like U^{88+}, B indicates a transition in B-like U^{87+}, C indicates a transition in C-like U^{86+}, and O indicates a transition in O-like U^{85+}. The key indicating the transition labels is given in Table II. The ^{235}U (^{233}U) spectrum is offset by 500 (950) counts/channel. The dashed lines indicate the position of the ^{238}U lines as determined by Ref. [13].

a MEVVA than using the platinum wire probe. The difference in the ionization balance for the different measurements does not affect the determination of bound-to-bound x-ray transition energies. The reason is that each of the observed lines is produced by a single transition from a single charge state, as discussed in detail in Ref. [21], and is not affected by satellite transitions from neighboring charge states that could shift the line centroid of the observed lines. The only blended feature is the B-like feature, which is formed by two nearly coincident transitions in B-like uranium. Like the other lines, however, this feature is not affected by satellite transitions from other charge states.

Measuring $\Delta E^{A,238}$ requires knowledge of the dispersion of the spectrometer, but an absolute calibration is unnecessary. To determine the dispersion, we employed the ^{238}U-transition-energy measurements of Ref. [13] (see Table II). The dispersion uncertainty from this procedure is 0.4%, which results in a 1-meV uncertainty in $\Delta E^{A,238}$. Because we are measuring energy differences between nearby lines, many systematic errors, such as detector nonlinearities or line shape uncertainties, cancel, permitting very precise measurements.

Data collection alternated between ^{235}U and ^{238}U spectra. By interleaving the spectra, we could monitor and correct for any possible electronic gain shifts. A similar procedure was followed earlier for the collection of the ^{233}U, ^{238}U data. The uncertainty associated with electronic gain drifts is approximately 5 meV.

TABLE III. Summary of the δE_{Coul} and the deduced $\delta\langle r^2\rangle^{235,238}$ values for each charge state. The uncertainties listed are entirely statistical.

Key	$\Delta E^{235,238}$ (meV)	δE_{Coul} (meV)	$\delta\langle r^2\rangle^{235,238}$ (fm^2)
Li	63±81	65±81	−0.085±0.106
Be	209±42	211±42	−0.284±0.056
B-1,2	200±38	202±38	−0.267±0.050
C	175±52	177±52	−0.236±0.070

ANALYSIS

Table III summarizes $\Delta E^{235,238}$ for each transition measured. For comparison, we list in Table IV the energy shift $\Delta E^{233,238}$ observed in Ref. [7]. Line fits to the $\Delta E^{235,238}$ data results in a slope of 15 ± 27 meV/charge and 32 ± 36 meV/charge to the $\Delta E^{233,238}$ data. These slopes are consistent with zero; that is, within the experimental uncertainty, ΔE is independent of the charge state. This finding is confirmed in a theoretical study of the effect of electron correlations on the transition energies. We calculated $\Delta E^{233,238}$ for the four ionization stages using a multiconfiguration Dirac-Fock (MCDF) [22] code and found differences no larger than 11 meV, affirming the small size of electron correlations. We performed a second calculation of the $\Delta E^{233,238}$ for the Li-like and Be-like transitions using a relativistic configuration interaction (RCI) code. The RCI calculations were done by increasing the basis set until convergence was achieved [23]. The results agreed within 0.1 meV with those from the MCDF calculations, affirming the predictive power of our calculations for ΔE and providing an uncertainty of less than 1 meV in the calculated size of the isotopic variation in the electron correlations.

In order to infer δE_{Coul} and thus $\delta\langle r^2\rangle$ from ΔE, we need to estimate the isotopic variation of the specific mass shift, of the QED terms, and of the nuclear polarization [16]. The advantage of our technique is that all these terms are small with correspondingly small uncertainties. The specific mass shift, also called the mass polarization contribution, has been calculated for the Li-like U^{89+} ion [24,25] and is similar for all ionization states under consideration here. It is found to be on the order of 50 meV with a theoretical uncertainty of 100% due to presently ignored terms of order $(Z\alpha)^2$. We estimate the isotopic variation of this value to be on the order of 1% (the mass difference between ^{233}U and ^{238}U) or less than 1 meV.

The estimate of the QED self-energy contribution to these energy levels is about 57 eV [25]. The finite nuclear size correction to this value is about 800 meV for ^{238}U. The

TABLE IV. Summary of the δE_{Coul} and the deduced $\delta\langle r^2\rangle^{233,238}$ values for each charge state. The uncertainties listed are entirely statistical.

Key	$\Delta E^{233,238}$ (meV)	δE_{Coul} (meV)	$\delta\langle r^2\rangle^{233,238}$ (fm^2)
Li	256±118	260±118	−0.338±0.153
Be	300±61	304±61	−0.409±0.081
B-1,2	320±52	324±52	−0.428±0.068
C	362±62	366±62	−0.488±0.083

TABLE V. Energy level and transition contributions due to nuclear polarization as given by Refs. [9,12]. The values noted with an asterisk are approximations based on linear extrapolation from the calculated values.

Isotope	ΔE (meV) $2s_{1/2}$	ΔE (meV) $2p_{1/2}$	ΔE (meV) $2p_{3/2}$	ΔE (meV) $2s_{1/2}$-$2p_{3/2}$	ΔE (meV) $2s_{1/2}$-$2p_{1/2}$
^{233}U			0	29.9*	
^{234}U	31.2	3.6	0	31.2	27.6
^{235}U			0	32.0*	
^{236}U	32.5	3.7	0	32.5	28.8
^{238}U	33.7	3.9	0	33.7	29.8

dependence of this value on the isotope can be estimated from Ref. [26] to be 4 meV (8 meV) for ^{235}U (^{233}U). The QED vacuum polarization contribution to the energy levels also has a nuclear size correction. The vacuum polarization contributes about -14 eV, and the nuclear size correction is about -760 meV [25]. Note that this value is almost equal and of opposite sign of the self-energy contribution. Thus, not only is the isotopic dependence of these two effects small, but they tend to cancel and can be ignored in the analysis of these data.

Nuclear polarization, or nuclear polarizability, calculations have been done for the $1s$, $2s$, and $2p$ levels in H-like U^{91+} ions for the even-A isotopes [9,12]. These calculations show a modest isotopic dependence, which must be taken into consideration in our data. Though it would be preferable if calculations existed also for the odd-A isotopes, we are forced to extrapolate the values of the even-A results to that for ^{233}U and ^{235}U. (Note that Refs. [4, 6] indicate that any even-odd staggering in this isotopic region is small compared to present experimental precision.) Since the entire correction for the singly excited $2s_{1/2}$-$2p_{3/2}$ transitions measured in this work comes from the $2s$ shell, the values calculated for the H-like U^{91+} $2s_{1/2}$ level accurately approximates that of all the charge states considered here. The nuclear polarization contribution difference between ^{235}U (^{233}U) and ^{238}U is 2 meV (4 meV). Table V shows the values of Refs. [9,12] along with our interpolations. The authors of Ref. [12] estimate the uncertainty in their calculations of the absolute size of the nuclear polarization contribution to be $\pm 25\%$. Thus we take the difference value also to be uncertain by 25%, or ± 1 meV.

This discussion shows that we only need to account for the nuclear polarization contribution to ΔE in order to determine the value of δE_{Coul} for each charge state. The results are summarized in Tables III and IV.

A final consideration must be given to the possibility of line shifts induced by the hyperfine interaction in a low-electron-density environment. This possibility was shown recently by Beiersdorfer *et al.* in the case of the $2s_{1/2}$-$2p_{3/2}$ transition in the three-electron Li-like Bi^{80+} ion [27]. The presence of a nuclear magnetic moment splits the $1s^2 2s_{1/2}$ ground level into two hyperfine components so that the $2s_{1/2}$-$2p_{3/2}$ transition actually consists of two transitions with different energies (0.8 eV apart in the case of ^{209}Bi). In the high-density or statistical limit, the average of the two transitions equals that of the $2s_{1/2}$-$2p_{3/2}$ transition in the absence of hyperfine splitting. In the low-density or collisionless limit, the lower of the two $1s^2 2s_{1/2}$ hyperfine levels is populated more than the upper level, resulting in a net shift of the energy of the averaged $2s_{1/2}$-$2p_{3/2}$ transition energy toward higher energies; i.e., on average a larger-than-statistical fraction decays to the lowest level, yielding a higher-energy x ray. For the case of Bi^{80+}, Beiersdorfer *et al.* showed that at electron densities of about 10^{12} cm^{-3}, i.e., at densities comparable to those in the present measurement, the intensity ratio of the two $2s_{1/2}$-$2p_{3/2}$ transitions differed by almost a factor of 2 from the statistical ratio [27]. Using a similar analysis as in Ref. [27], we find, however, that this effect is negligible for ^{235}U. The splitting of the two $2s_{1/2}$-$2p_{3/2}$ hyperfine components in ^{235}U is 0.124 eV. However, because ^{235}U has a much smaller nuclear magnetic moment than ^{209}Bi—$0.35\mu_n$ versus $4.08\mu_n$—the density threshold at which substantial deviations from the statistical limit occur is lowered by more than two orders of magnitude to about 10^{10} cm^{-3}. The present measurements were, thus, performed near the high-density limit. At a density of 10^{12} cm^{-3}, we estimate that the measured $2s_{1/2}$-$2p_{3/2}$ transition energy differs by no more than 1 meV from its nominal statistical value.

The contributions from systematic experimental errors to the overall uncertainty of each δE_{Coul} value, as well as those arising from uncertainties in the atomic calculations, are summarized in Table VI. As seen from the table, the systematic uncertainties are small compared to the statistical uncertainties of our measurements. Gain drifts represent the largest contribution to the overall systematic uncertainty. This contrasts with our previous analysis [7] where the systematic uncertainty was dominated by the uncertainty in the theoretical determination of the nuclear polarization contribution. The change was brought about by the recent order-of-magnitude reduction in the size of the calculated nuclear polarization contribution [9] and, thus, an order-of-magnitude

TABLE VI. Summary of the contributions to the systematic uncertainties associated with δE_{Coul}. All are small compared to the statistical uncertainties (see Table I).

Contribution	$A=233$	$A=235$
Isotopic enrichment	<1 meV	<1 meV
Dispersion	1 meV	1 meV
Gain drifts	5 meV	5 meV
Mass polarization	<1 meV	<1 meV
Self-energy, vacuum polarization	1 meV	1 meV
Nuclear polarization	1 meV	<1 meV
Quadrature sum	5 meV	5 meV

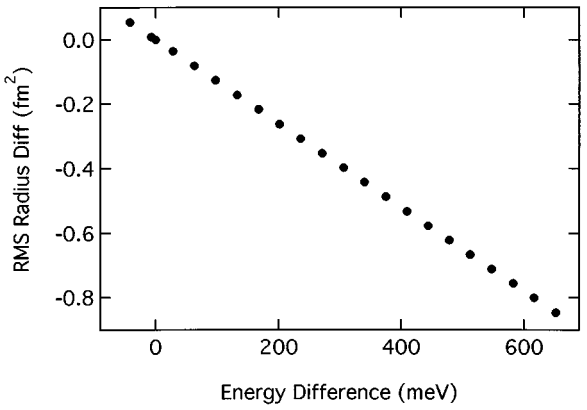

FIG. 2. Calculated values of $\delta\langle r^2\rangle$ vs δE_{Coul} for the $2s_{1/2}$-$2p_{3/2}$ transition in the Li-like charge state in U.

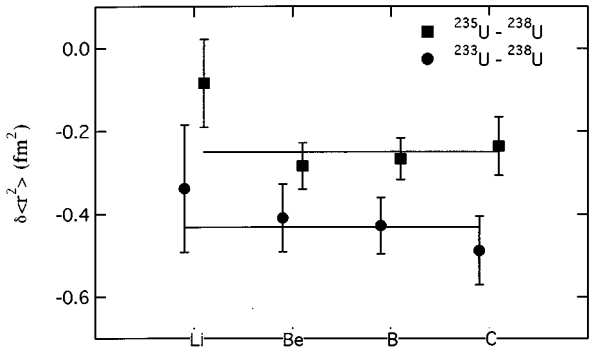

FIG. 3. Measured $\delta\langle r^2\rangle^{A,238}$ as a function of ion charge state. The solid lines represent the weighted average of the four transitions for each isotope pair.

reduction in the size of the associated uncertainty. The much smaller size in the nuclear polarization contribution can also be noted when comparing the values of δE_{Coul} inferred for the ^{233}U and ^{238}U pair with those listed in Ref. [7].

NUCLEAR SIZE DETERMINATIONS

The MCDF [22] calculations use a nuclear charge density function $\rho(r)$ described by the two-parameter Fermi distribution

$$\rho(r)=\rho_0/(1+e^{[r-\mu]/\tau}),$$

where r is the radius, μ is the half-density radius, and τ is the skin thickness. The resultant energy level determinations, however, are not sensitive to the actual charge distribution provided that the associated root mean square radius (r_{rms}) is reproduced. We calculated δE_{Coul} for each charge state for 22 values of μ between 7.038 11 and 7.143 95 fm, holding τ constant, and computed the corresponding r_{rms}. Figure 2 shows the results of these calculations for the case of the Li-like charge state. The results of these calculations provide $\delta\langle r^2\rangle^{A,238}$ as a function of δE_{Coul} for each ionization stage. The origin is defined as the values for ^{238}U ($\mu=7.137\,53$ fm and $\tau=0.523\,39$ fm), which correspond to a two-parameter Fermi distribution with $r_{\mathrm{rms}}=5.8610$ fm. This r_{rms} is equal to the value one derives from a four-parameter deformed-Fermi distribution using the parameters given in Ref. [6].

To deduce $\delta\langle r^2\rangle^{A,238}$ from δE_{Coul} using the curves described above, we did a quadratic interpolation between the calculated points for each charge state. The results for each of the lines measured in $A=235$ are listed in Table III. A summary of the $\delta\langle r^2\rangle^{235,238}$ results for the different charge states is shown in Fig. 3. Averaging the results, we find $\delta\langle r^2\rangle^{235,238}=-0.250$ fm^2 with a statistical uncertainty of 0.031 fm^2. The procedure of deducing $\delta\langle r^2\rangle$ for each charge state separately and then averaging ensures proper treatment of the electron correlation contribution. The systematic uncertainty in δE_{Coul} (5 meV) translates into a systematic uncertainty in $\delta\langle r^2\rangle^{235,238}$ of 0.006 fm^2. Adding the uncertainties in quadrature, the final result is $\delta\langle r^2\rangle^{235,238}=-0.250\pm0.032$ fm^2.

The results for $\delta\langle r^2\rangle^{233,238}$ of each of the measured lines in $A=233$ are listed in Table IV and are plotted in Fig. 3. The values are somewhat smaller than those given in Ref. [7], because of the revised nuclear polarization contributions from Ref. [9]. Averaging the results, we find $\delta\langle r^2\rangle^{233,238}=-0.432$ fm^2 with a statistical uncertainty of 0.042 fm^2. The systematic uncertainty in δE_{Coul} (5 meV) translates into a systematic uncertainty in $\delta\langle r^2\rangle^{233,238}$ of 0.006 fm^2. Adding the uncertainties in quadrature, the final result is $\delta\langle r^2\rangle^{233,238}=-0.432\pm0.043$ fm^2. This value is about 5% smaller than that of Ref. [7], because of the above-mentioned revision of the nuclear polarization contributions in Ref. [9].

The authors of Ref. [4] used the $K\alpha$ value for $\delta\langle r^2\rangle^{233,238}$ [5] to calibrate isotopic frequency shifts ($\delta\nu$) in terms of $\delta\langle r^2\rangle^{A,238}$. We performed a similar procedure, but used our two $\delta\langle r^2\rangle$ values to calibrate the $\delta\nu$ values. The results of this calculation are given in Table VII. The deduced constant to convert $\delta\nu^{233,238}$ to $\delta\langle r^2\rangle^{233,238}$ is $-3.27\pm0.24\times10^{-5}$ fm^2/MHz. The deduced constant to convert $\delta\nu^{235,238}$ to $\delta\langle r^2\rangle^{235,238}$ is $-2.95\pm0.38\times10^{-5}$ fm^2/MHz. The weighted average of these two constants is $-3.16\pm0.20\times10^{-5}$ fm^2/MHz. Using two calibration points results in a lower uncertainty as compared to the previous determination based on a single $K\alpha$ measurement. The value of $\delta\langle r^2\rangle^{233,238}$ determined in this way is -0.416 ± 0.028 fm^2. That for $\delta\langle r^2\rangle^{235,238}$ is -0.268 ± 0.017 fm^2. Table VII summarizes the optical values treated in this manner, and Fig. 4 shows a summary of all the U $\delta\langle r^2\rangle$ measurements under discussion here.

Our result for $\delta\langle r^2\rangle^{233,238}$ can be compared with that of previous studies: -0.383 ± 0.044 fm^2 [4,5] and -0.520

TABLE VII. Values of $\delta\langle r^2\rangle^{A,238}$ derived from the isotopic frequency shift ($\delta\nu$) results of Ref. [4] using the $\delta\langle r^2\rangle^{233,238}$ and $\delta\langle r^2\rangle^{235,238}$ results from the current work as a calibration. The $\delta\nu^{A,238}$ values are those associated with the 5915.42-Å $5f^36d7s^2\ {}^5L_6^0$-$5f^36d7s7p\ {}^7M_7$ transition in neutral uranium.

A	$\delta\nu^{A,238}$ (MHz)	$\delta\langle r^2\rangle^{A,238}$ (fm^2)
233	13174.0±3.1	−0.416±0.026
234	10108.5±4.2	−0.319±0.020
235	8480.5±3.6	−0.268±0.017
236	5069.5±4.8	−0.160±0.010

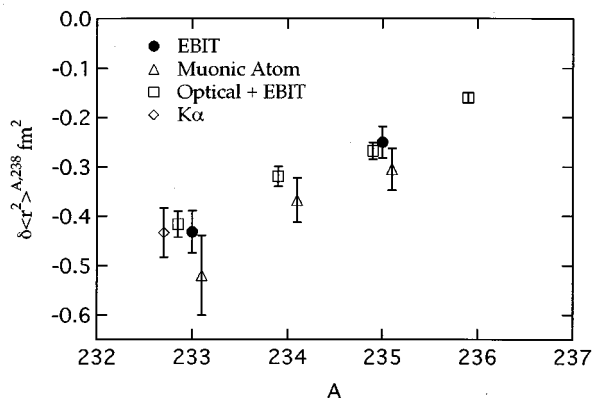

FIG. 4. Summary of the measurements of $\delta\langle r^2\rangle^{A,238}$ in the U system. The optical data were normalized as described in the text using the current results as the normalization.

± 0.081 fm^2 [6]. However, Istvan [28] pointed out that although the authors of Ref. [4] quoted their result as a value for $\delta\langle r^2\rangle^{233,238}$, it is actually the value for the λ parameter defined as

$$\lambda = \delta E_{\mathrm{Coul}}/C_1 = \delta\langle r^2\rangle + (C_1/C_2)\,\delta\langle r^4\rangle + \cdots ,$$

where C_1 and C_2 are the Seltzer coefficients [28]. Although λ and $\delta\langle r^2\rangle$ are similar, they differ by approximately 10% for $\delta\langle r^2\rangle^{233,238}$. Using the Seltzer coefficients [29] for U ($C_1 = 4700$ meV/fm^2, $C_2 = -6.01$ meV/fm^2), one can determine that the correct value to use for $\delta\langle r^2\rangle^{233,238}$ from Ref. [4] by iteration of the above formula. The result is -0.433 ± 0.050 fm^2. The weighted mean of all measurements is -0.445 ± 0.030 fm^2. All three experiments are consistent with this mean value to within one to two standard deviations.

DISCUSSION

We have performed high-precision spectroscopy on radioactive few-electron highly charged uranium ions trapped in an EBIT in order to determine the variation in nuclear charge radii between different isotopes. Our measurement focused on the isotopic energy shift between ^{235}U and ^{238}U for the $2s_{1/2}$-$2p_{3/2}$ transitions in U^{86+} through U^{89+} ions. The advantage of employing such highly charged ions for these measurements lies in the fact that the atomic structure of few-electron ions is relatively simple and can be calculated with a high degree of accuracy. This is especially true, because our measurements are concerned with energy shifts, where the absolute transition energy is of little importance. Contributions from the specific mass shift, which can dominate the analysis of spectral data from neutral or few-times ionized atoms, are unimportant. The measured energy shift in our study is within about 1% directly caused by the change in the nuclear charge radius. Extracting the variation in the nuclear charge radii is thus intuitive and relatively uncomplicated, and the uncertainties associated with the analysis procedure are accordingly small.

The uncertainty of our current measurement is lower than that of the earlier measurement using muonic atoms [6]. As a result, we can improve on earlier determinations of the nuclear radii variations of isotopes that have not been measured directly. Combining our present result for $\delta\langle r^2\rangle^{235,238}$ with our earlier measurement of $\delta\langle r^2\rangle^{233,238}$, we have been able to reanalyze the optical data from Ref. [4] and have obtained improved values for $\delta\langle r^2\rangle^{A,238}$ with $A = 233$, 234, 235, and 236. A further improvement may, in principle, be achieved by performing an average over all measured values, i.e., by averaging our results, those from muonic atoms and those from $K\alpha$ measurement of neutral uranium. Such an average is suggested by the fact that most measurements overlap within their respective uncertainty limits and appear to be statistically distributed.

The uncertainties of our measurement are virtually all from statistical considerations. In other words, the uncertainties are dominated by the uncertainty with which the line centroids could be determined, which in turn is related to the number of counts in a given line and its width. In the present measurement, the measured shift is found to be about 20% of the observed linewidth. The width may be reduced by employing crystal spectrometers with yet better resolving power. Spectrometers with resolving powers as high as 68 000 have been employed successfully to study the ion temperature by observing the Doppler-broadened line profiles in various highly charged ions [30,31]. These studies have shown that the ion temperature can be reduced to values below 100 eV. Under these circumstances, the linewidth would be reduced by about a factor of 5 over that of the present measurement. Correspondingly, a 5 times better measurement of the nuclear charge radius variation could be achieved. While no such measurements have yet been performed, they point to the level of precision that could be achieved in spectroscopic studies of highly charged ions for the purpose of determining nuclear structure parameters in the future. This precision would lead to yet better determinations of the isotopic dependence of the nuclear charge radii of heavy isotopes.

ACKNOWLEDGMENTS

We are grateful for the technical expertise and assistance of E. W. Magee and R. Lougheed in the preparation and handling of the wire probes. This work was performed under the auspices of the U.S. Department of Energy at Lawrence National Laboratory under Contract No. W-7405-Eng-48.

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