Trapped-Ion Technique for Measuring the Nuclear Charge Radii of Highly Charged Radioactive Isotopes

S. R. Elliott
Department of Physics, University of Washington, P.O. Box 351560, Seattle, Washington 98195-1560

P. Beiersdorfer and M. H. Chen
Department of Physics and Space Technology, Lawrence Livermore National Laboratory, Livermore, California 94550
(Received 21 August 1995)

We present the first measurement to isolate the variation of nuclear effects in the x-ray transitions of few-electron heavy ions. Using a novel technique to produce and trap radioactive ions we measured the energy difference between $2s_{1/2}$-$2p_{3/2}$ transitions in Li-, Be-, B-, and C-like ^{233}U and ^{238}U. We show that because of the simplified atomic structure of few-electron ions the data are readily interpreted in terms of the variation in the mean nuclear radius. A value $\delta\langle r^2\rangle^{233,238} = -0.457 \pm 0.043$ fm^2 is found, which lies between earlier measurements based on different techniques.

PACS numbers: 21.10.Ft, 27.90.+b, 29.25.Ni, 32.30.Rj

The trapping of radioactive nuclides creates new opportunities for novel nuclear physics experiments. The recent success of trapping radioactive neutral atoms [1,2], for example, has opened the possibility to sensitive beta decay asymmetry studies. Low-charged ions, held in a Penning trap [3] or Paul trap [4], have provided valuable data on the masses of unstable isotopes. In this Letter, we report the first nuclear physics measurements using trapped few-electron, very-high-Z radioactive ions. Using precision x-ray spectroscopy and exploiting the simplified electronic structure of few-electron ions we isolate the nuclear effects among different isotopes and infer the isotopic variation of the nuclear charge distribution, a fundamental parameter crucial for understanding the collective structure of the nucleus (see, for example, Refs. [5–8]). Its variation, parametrized in terms of the change in mean-square nuclear charge radius ($\delta\langle r^2\rangle$), has been inferred in the high-Z region from muonic-atom x rays [9] and neutral-atom optical isotope shift studies [10]. Our present measurement focuses on the isotopes ^{233}U and ^{238}U for which earlier measurements of $\delta\langle r^2\rangle$ have produced discrepant results, i.e., -0.520 ± 0.081 fm^2 [11] and -0.383 ± 0.044 fm^2 [12,13].

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 was previously impossible because of the lack of a facility at which the x-ray transitions from such highly stripped radioactive ions could be generated and measured conveniently and reliably. This situation has changed recently with the successful implementation of a high-energy electron beam ion trap [14] that allows the production of very highly charged ions and of efficient crystal spectrometers [15] that can resolve individual transitions with very high resolution. Moreover, ion traps generally require only minute quantities of material for filling. Thus they are well suited for investigating the properties of isotopes that are rare or radioactive. The transitions studied in the present measurement 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 I lists the specific transitions studied and their energies in ^{238}U measured in Ref. [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,

TABLE I. Summary of the measured energy shifts. The ^{238}U energy values and the nomenclature for the key are from Ref. [15]. All transitions decay to the ground state of the respective ion.

Key	Ion	Upper level	^{238}U energy (eV)	ΔE (meV) ^{233}U-^{238}U
Li	U^{89+}	$(2p_{3/2})_{j=3/2}$	4459.37 ± 0.35	256 ± 118
Be	U^{88+}	$(2s_{1/2}2p_{1/2})_{j=1}$	4501.72 ± 0.27	300 ± 61
B-1,2	U^{87+}	$(2s_{1/2}2p_{1/2}2p_{3/2})_{j=1/2,3/2}$ blend	4521.39 ± 0.22	320 ± 52
C	U^{86+}	$(2s_{1/2}2p_{1/2}^2 2p_{3/2})_{j=1}$	4548.32 ± 0.20	362 ± 62
O-1	U^{84+}	$(2s_{1/2}2p_{1/2}^2 2p_{3/2}^3)_{j=2}$	4525.26 ± 0.25	

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 atoms [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. 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 [14]. An electron beam ionizes, excites, and radially traps the ions. The ions are trapped axially by potential differences between three collinear cylindrical electrodes through which the beam passes. Low-charged ions, injected into the trap, are ionized to high charge states by successive collisions with beam electrons.

The ^{233}U ions were introduced into the trap using a novel method [17] relying on a thin wire platinum probe with a plated tip placed near the electron beam. The total mass of plated ^{233}U, isotopically enriched to 99.92%, was only 100 ng and less than 10 ng were consumed during the course of the experiment. The ^{238}U ions were provided by a standard metal vapor vacuum arc source [18] using a ^{238}U cathode depleted in ^{235}U weighing 14 g.

The ions are 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 [19]. The spectrometer uses 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 are recorded with a gas-filled position sensitive proportional counter with a $10 \times 3 \times 1$ cm^3 active volume. The energy resolution of the setup was 1.1 eV FWHM, i.e., more than 2.5 times better than the measurement in Ref. [15].

The x-ray spectrum of ^{233}U was compared with that from ^{238}U. Figure 1 shows the two measured spectra. Each spectrum was accumulated over 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- and B-like ionization stages in the ^{233}U data than for the ^{238}U data. Thus the ^{238}U spectrum shows a weak O-like line which is absent in the ^{233}U data.

Measuring $\Delta E^{233,238}$ 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. [15] (see Table I). The dispersion uncertainty from this procedure is 0.4% which results in a 1 meV uncertainty in

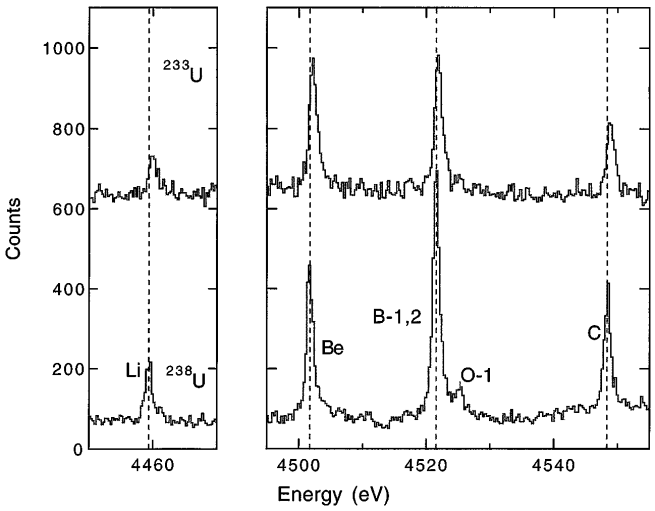

FIG. 1. Crystal spectrometer spectra of the $2s_{1/2}$-$2p_{3/2}$ transitions in U^{85+} through U^{89+} for the two isotopes ^{233}U and ^{238}U. The key indicating the transition labels is given in Table I. The ^{233}U spectrum is offset by 500 counts/channel. The dashed lines indicate the position of the ^{238}U lines as determined in Ref. [15].

$\Delta E^{233,238}$. Because we are measuring energy differences between nearby lines, many systematic errors, such as detector nonlinearities, cancel permitting very precise measurements.

Data collection alternated between ^{233}U and ^{238}U spectra. By interleaving the spectra, we could monitor and correct for any possible electronic gain shifts. No correction was required and the uncertainty associated with electronic gain drifts is approximately 5 meV.

Table I summarizes ΔE for each transition measured. The contributions from systematic errors to the overall uncertainty of each ΔE value are small and are summarized in Table II. A line fit to these ΔE data results in a slope ($m = 32 \pm 36$ meV/charge) which is consistent with zero; that is, ΔE is nearly 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

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

Contribution	Uncertainty
Isotopic enrichment	<1 meV
Dispersion	1 meV
Gain drifts	5 meV
Mass polarization	<1 meV
Self-energy, vacuum polarization	1 meV
Nuclear polarization	6 meV
Quadrature sum	8 meV

ionization stages using a multiconfiguration Dirac-Fock (MCDF) [20] code and found differences no larger than 11 meV, affirming the small size of electron correlations. We performed a second calculation of $\Delta E^{233,238}$ for the Li-like and Be-like transitions using a relativistic configuration interaction (RCI) code with B-spline basis sets. The RCI calculations were done by increasing the basis set until convergence was achieved [21]. 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 [22,23] and is similar for all ionization states under consideration here. It is found to be of 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 and vacuum polarization contributions to these energy transitions is about 45 eV [23]. The finite nuclear size corrections to these contributions are each about 800 meV, but are of opposite sign so that the sum vanishes. The isotopic variation of each contribution is approximately 8 meV, and also tends to cancel in the sum and thus can be ignored in the present analysis.

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 [24]. 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. (Note that Refs. [11,12] 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 ^{233}U and ^{238}U is 24 meV. The authors of Ref. [24] 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 ± 6 meV. Eliminating the nuclear polarization contribution to ΔE yields a final value for δE_{Coul} for each charge state as summarized in Table III.

The MCDF [20] 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}. 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. [11].

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. These results, listed in Table III, were then averaged and we find $\delta\langle r^2\rangle^{233,238} = -0.457$ fm^2 with a statistical uncertainty of 0.042 fm^2. This 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} (8 meV) translates into a systematic uncertainty in $\delta\langle r^2\rangle^{233,238}$ of 0.010 fm^2. Adding the uncertainties in quadrature, the final result is $\delta\langle r^2\rangle^{233,238} = -0.457 \pm 0.043$ fm^2. This result can be compared with that of previous studies: -0.383 ± 0.044 [12,13] and -0.520 ± 0.081 fm^2 [11]. The present measurement thus favors neither of the earlier measurements. The weighted mean of all measurements is -0.434 ± 0.028 fm^2. All three experiments are consistent with this mean value to within 1 to 2 standard deviations.

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

Key	δE_{Coul} (meV)	$\delta\langle r^2\rangle^{233,238}$ (fm^2)
Li	280 ± 118	-0.364 ± 0.153
Be	324 ± 61	-0.436 ± 0.081
B-1,2	344 ± 52	-0.455 ± 0.068
C	386 ± 62	-0.515 ± 0.083
Average		-0.457 ± 0.043
Previous	$K\alpha$ [12,13]	-0.383 ± 0.044
	Muonic atoms [11]	-0.520 ± 0.081

In summary, we have performed the first nuclear measurements on radioactive few-electron highly charged uranium ions trapped in an EBIT. Our measurement demonstrates that trapped-based nuclear physics can be extended from neutral or one- or few-times ionized atoms in Zeeman, Paul, or Penning traps to very highly charged ions in electron beam ion traps. Because only minute quantities of isotopically pure material were needed, our technique should readily be applicable to a wide variety of rare or radioactive isotopes. The present measurements represent the first time precise isotope shift measurements have been made in electronic transitions that are strongly affected by quantum electrodynamics. In fact, the uncertainties in the nuclear properties are the dominant constraints on the accuracy with which the quantum electrodynamical contributions to the transition energies can be determined. The spread of 0.137 fm^2 in $\delta\langle r^2\rangle$ in the previous measurements of the nuclear radii [11–13] corresponds to an uncertainty in the transition energies of 0.10 eV. This uncertainty previously has been ignored in QED studies. That is, the theoretical calculations have used the muonic x-ray results for the charge radius without commenting on the discrepancy with the $K\alpha$ results for $\delta\langle r^2\rangle$. The present measurement reduces this uncertainty to 0.03 eV. The current results are statistically limited. A precision limited by the uncertainties in the nuclear polarization calculations can in principle be achieved. By choosing isotopic systems where $\delta\langle r^2\rangle$ is very well known, experimental tests of the nuclear polarization calculations might soon be possible.

We are grateful for the technical expertise and assistance of E. W. Magee and R. Lougheed. Discussions with Mort Weiss are greatly appreciated. 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.

[1] Z.-T. Lu *et al.,* Phys. Rev. Lett. **72**, 3791 (1994).
[2] G. Gwinner *et al.,* Phys. Rev. Lett. **72**, 3795 (1994).
[3] H. Stolzenberg *et al.,* Phys. Rev. Lett. **65**, 3104 (1990).
[4] W. Kalber *et al.,* Z. Phys. A **334**, 103 (1989); J. Rink *et al.,* Z. Phys. A **342**, 487 (1992).
[5] Z. Lojewski *et al.,* Phys. Rev. C **51**, 601 (1995).
[6] Bozena Nerlo-Pomorska, Krzysztof Pomorski, and Beata Skorupska Mach, Nucl. Phys. **A562**, 180 (1993).
[7] B. Q. Chen and P. Vogel, Phys. Rev. C **48**, 1392 (1993).
[8] Uwe Regge and Dietrich Zaischa, Comments At. Mol. Phys. **23**, 257 (1989).
[9] R. Engfer *et al.,* At. Data Nucl. Data Tables **14**, 509 (1974).
[10] P. Aufmuth, K. Heilig, and A. Steudel, At. Data Nucl. Data Tables **37**, 455 (1987).
[11] J. D. Zumbro *et al.,* Phys. Rev. Lett. **53**, 1888 (1984).
[12] A. Anastassov *et al.,* Hyperfine Interact. **74**, 31 (1992).
[13] R. T. Brockmeier, F. Boehm, and E. N. Hatch, Phys. Rev. Lett. **15**, 132 (1965).
[14] D. A. Knapp *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. A **334**, 305 (1993).
[15] P. Beiersdorfer *et al.,* Phys. Rev. Lett. **71**, 3939 (1993).
[16] W. H. King, *Isotope Shifts in Atomic Spectra* (Plenum Press, New York, 1984).
[17] S. R. Elliott and R. E. Marrs, Nucl. Instrum. Methods Phys. Res., Sect. B **100**, 529 (1995).
[18] I. G. Brown *et al.,* Appl. Phys. Lett. **49**, 1019 (1986).
[19] P. Beiersdorfer *et al.,* Rev. Sci. Instrum. **61**, 2338 (1990).
[20] I. P. Grant *et al.,* Comput. Phys. Commun. **21**, 207 (1980); B. J. McKenzie *et al., ibid.* **21**, 233 (1980).
[21] M. H. Chen, K.-T. Cheng, and W. R. Johnson, Phys. Rev. A **47**, 3692 (1993).
[22] S. A. Blundell, W. R. Johnson, and J. Sapirstein, Phys. Rev. A **41**, 1698 (1990).
[23] S. A. Blundell, Phys. Rev. A **46**, 3762 (1992).
[24] Guenter Plunien *et al.,* Phys. Rev. A **43**, 5853 (1991); G. Plunien and Gerhard Soff, *ibid.* **51**, 1119 (1995).