

Determination of nuclear charge radius by extreme-ultraviolet spectroscopy of Na-like ions

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We report on a method for determining the absolute nuclear charge radius of high- Z elements using extreme-ultraviolet spectroscopy of highly charged Na-like ions in tandem with highly accurate atomic structure calculations of transition energy differences. The application of this method has reduced the nuclear charge radius uncertainty of ^{191}Ir by a factor of 8 from the currently accepted literature value, with a recently reported charge radius of 5.435(12) fm. The result reduces the charge radius uncertainty along the full Ir isotopic chain when combined with prior optical isotope shift measurements. The technique utilizes only a few million ions stored in an ion trap, which should apply to measurements with small quantities of radioactive nuclei.

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Introduction. The nuclear charge radius is a fundamental property that is needed in a variety of investigations, such as nuclear structure [1], shape and odd-even staggering [2–6], nuclear halos [7,8], shape coexistence [9], atomic parity violation and searches for permanent electric dipole moments [10,11], strong-field quantum electrodynamics (QED) tests [12], and searches for physics beyond the standard model [13]. Experimental techniques that can accurately determine the nuclear charge radius are needed to aid these studies.

Muonic atom x-ray spectroscopy [14–16] and elastic electron scattering [15,17,18] are the only main traditional methods to measure $R = \langle r^2 \rangle^{1/2}$, the root-mean-square (rms) radius of a nuclear charge distribution [19]. These techniques are typically applied to stable nuclei due to intrinsic experimental challenges (e.g., a few μg of target material is needed for the experiments). Except for the SCRIT ion trap facility that has only recently extended elastic electron scattering to radioactive nuclei [20], no other technique is currently available to measure the absolute nuclear charge radii with $<10^8$ nuclei available.

Consistent sets of nuclear charge radii have been compiled by several groups [19,21,22]. The latest critically evaluated data [19] were constructed from combined absolute nuclear charge radii constrained by measurements of relative charge radii from $K\alpha$ isotope shift [23,24] and precise optical isotope shift (OIS) [25–29] experiments. Despite efforts to determine accurate nuclear charge radii, absolute experimental determinations are scarce or have large uncertainties.

We present a method to determine the absolute nuclear charge radius of heavy elements by utilizing a nearby element with a well-known nuclear charge radius as a reference. The technique relies on precise measurement of the separation of the Na-like $D1\ 3s\ ^2S_{1/2}-3p\ ^2P_{1/2}$ transitions of two elements, coupled with highly accurate atomic structure calculations of the same.

We describe a measurement where the previously recommended uncertainty [19] of the nuclear charge radius of ^{191}Ir is reduced by a factor of 8. Efforts are under way to determine the nuclear charge radius of rare nuclei that have no stable isotopes and are only available at radioactive isotope beam facilities [30].

Nuclear charge radius from Na-like ions. The $3s-3p$ resonance transition in single valence electron Na-like ions uniquely facilitates investigations of nuclear effects on atomic properties [31]. Highly charged Na-like ions exhibit a compressed electron cloud, enhancing the importance of nuclear, relativistic, and QED effects, and can be comfortably produced experimentally in small-scale laboratory devices.

Unlike in other more complex atomic systems, modern atomic structure theory can now accurately calculate these

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effects in Na-like ions, including fundamental electron-electron interactions. The sensitivity to the nuclear charge radius is generally limited by omitted terms in the calculations. Single valence electron configurations in general (e.g., H- and Li-like systems) exhibit similar nuclear charge radius sensitivity [32]; however, these charge states are difficult to create experimentally in heavy ions, as they require higher electron beam energy super-electron beam ion trap (EBIT) devices.

The key point to our method based on Na-like ions is that the omitted QED and many-body corrections—limiting the theoretical uncertainties of the absolute transition energies—largely cancel out in transition energy difference calculations. These terms can be described by a smooth function of Z and can be estimated from Z -dependent theoretical uncertainties [31]. Although nuclear mass terms will give nonsmooth contributions as a function of Z , these terms are relatively small in heavy ion Na-like transitions [31].

The technique is based on the comparison of the measured and calculated Na-like D1 $3s\ 2S_{1/2}-3p\ 2P_{1/2}$ transition energy difference between two nearby isoelectronic ions $\delta E_{AB} \equiv E_{A-B}^{\text{exp}} - E_{A-B}^{\text{th}}$. If the nuclear radius of one of them is precisely known (R_A), the mismatch between theory and experiment can be expressed using the linear nuclear sensitivity coefficient of the other [31]:

$$\delta E_{AB} = S_B \delta R_B. \quad (1)$$

Here, $\delta R_B \equiv R_B - R_{B_0}$, and S_B is the nuclear sensitivity coefficient of nuclide B . Also, R_{B_0} is the charge radius used in the calculations, which is assumed to differ from the R_B real value by δR_B . The linearity condition requires that $\delta R_B \ll R_{B_0}$.

With an accurate nuclear sensitivity coefficient and precisely measured and calculated energy differences, one can use Eq. (1) to derive δR_B and, accordingly, the absolute nuclear charge radius of nuclide B as $R_B = R_{B_0} + \delta R_B$.

Experimental details. The EBIT at the National Institute of Standards and Technology (NIST) [33] used a 2.7 T magnetic field to compress a 150 mA, 18 keV electron beam to create Na-like charge states of Os and Ir injected from a metal Vvapor vacuum arc external ion source [34].

In these measurements, natural abundance Os and Ir sample materials were used; however, for both elements, high-precision differences in isotopic nuclear charge radii can be found in the literature [19] because of precise OIS measurements available for both Os and Ir [27,35–37].

To assess mass-dependent processes in the EBIT (e.g., electron-beam heating, ion-ion collisions, and evaporative cooling) that could potentially alter the natural isotopic abundance ratios of ions in higher charge states, we have conducted a measurement at the EBIT of the Facility for Rare Isotope Beams [38].

In this setup, an extraction beamline was available for the direct measurement of ion currents of different isotopes and charge states of Ne. Experiments with highly ionized H-like ^{22}Ne and ^{20}Ne showed that the isotope ratios reproduced the precise literature value [39] within 2%, with a measurement uncertainty of 1%. Simulations reproducing these results indicate that this effect is negligible for much heavier ions (almost 10 times the mass) and does not affect the current Ir-Os measurements.

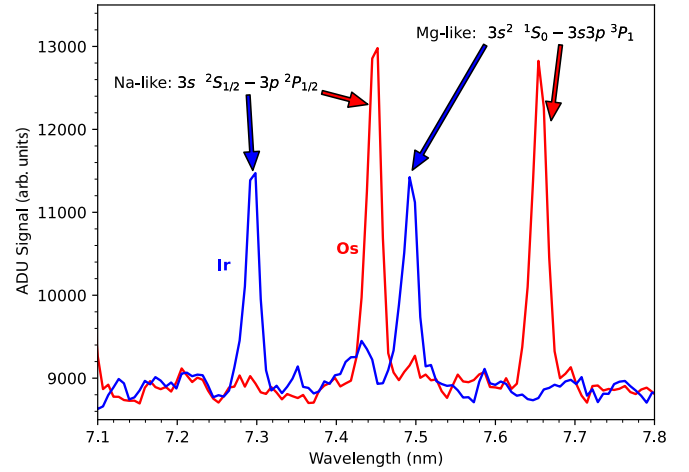


FIG. 1. Experimental extreme-ultraviolet (EUV) D1 lines of Na-like Os (red) and Ir (blue) in the NIST electron beam ion trap (EBIT).

The 4 nm to 20 nm extreme-ultraviolet (EUV) range radiation, from an estimated few million ions confined within the EBIT trap region, was recorded by a flat-field grazing incidence spectrometer, equipped with a liquid nitrogen cooled charge-coupled device camera [40]. Ne, Xe, and Ar emission lines were used to calibrate the spectrometer [41]. A total of 262 spectra alternating between Os, Ir, and calibration elements were recorded over 4 days. Cosmic ray filtering was performed by evaluating the Poisson noise between sequential frames of spectra [41]. Figure 1 shows a portion of the spectra of highly charged ions of Os and Ir within this range, highlighting the spectral features observed during the experiment.

Theoretical calculations. The calculations of the atomic structure of Na-like Os and Ir ions made use of two methods: the fully relativistic multiconfiguration Dirac-Hartree-Fock (MCDHF) method (GRASP2K [42]) and relativistic many-body perturbation (RMBPT [43]). The MCDHF method is based on the Dirac-Coulomb-Breit Hamiltonian that includes correlations and Breit contributions, assuming a two-parameter Fermi nuclear charge distribution. For QED corrections, GRASP2K includes the one-loop self-energy and vacuum polarization contributions using a screened hydrogenic wave function. However, higher-order QED effects can be prominent for high- Z elements and need to be considered. In this letter, we adjusted the QED corrections computed by GRASP2K using values obtained from the *ab initio* QED treatment given in Ref. [31]. MCDHF calculations explicitly consider isotope specific nuclear charge radii (field shifts) and include recoil (mass) corrections, hyperfine effects, as well as nuclear deformations. Calculations using RMBPT were updated in Ref. [31] with *ab initio* QED corrections from Ref. [44]. The RMBPT calculations for the transition energies of isotopes considered mass shifts and field shifts as well, using the mass-shift coefficient and nuclear-sensitive coefficient, respectively [31].

Hyperfine splitting of lines for nuclei with nonzero nuclear magnetic moments has been investigated by calculating relative intensities using collisional-radiative kinetics of hyperfine levels with the NOMAD [45] code for non-Maxwellian plasma. This effect was negligible for the current precision but can become significant with the improvement of the method.

TABLE I. Experimental and theoretical quantities used in Eq. (2) to calculate the extracted charge radius correction.

Parameter	Value
$E_{\text{Ir-Os}}^{\text{exp}} \text{ (cm}^{-1}\text{)}$	28 249 (35)
$E_{\text{Ir-Os}}^{\text{RMBPT}} \text{ (cm}^{-1}\text{)}$	28 387 (10)
$E_{\text{Ir-Os}}^{\text{MCDHF}} \text{ (cm}^{-1}\text{)}$	28 396 (10)
$S_{\text{Ir}}^{\text{RMBPT}} \text{ (cm}^{-1}\text{/fm)}$	−3723 (186)
$S_{\text{Ir}}^{\text{MCDHF}} \text{ (cm}^{-1}\text{/fm)}$	−3721 (186)

The dominant uncertainty in S arises from nuclear-model dependence, which we estimate to be $\sim 4\%$ of S . This estimate is based on evaluating S for a range of plausible nuclear models (including models with and without deformation) and noting fluctuations in values obtained. The 4% uncertainty also accounts for the effects of nonlinearities beyond the linear approximation in Eq. (1). Nuclear polarization is not considered in this letter; however, authors of previous studies on Li-like ions [46] have shown that its effect is of similar magnitude to deformation. To account for this missing contribution, we have included an additional 1% in our uncertainty budget for the nuclear sensitivity coefficient S , which thus became 5% overall.

Our theoretical calculations using the MCDHF and RMBPT methods consistently provide the same values for the transition energy differences E_{A-B}^{th} within their respective uncertainties, even for cases where the predicted transition energies are different. Furthermore, transition energy differences of the S -matrix method by Sapirstein and Chen [47] are consistent with these values. The uncertainty of the difference is much smaller than the uncertainties of the transition energies themselves; therefore, the agreement provides confidence that these high-precision calculations can be used for this technique.

The separation of the Na-like D1 lines was determined through two methods: (1) the absolute difference of the measured line centers based on all available Ir and Os spectra and (2) the directly measured difference of the line positions by comparing time-adjacent pairs of repeated Ir and Os measurements with a single fitted separation [49]. Monte Carlo simulations that varied the D1 and calibration line centers were used to estimate the correlation [50] between the two methods, allowing for the combination of the experimental separation given in Table I.

The calculated transition energy difference between Ir and Os in Table I was obtained by weighted averaging the transition energies of the different isotopes, considering the accurately known nuclear radius separations [19,27,35–37] and the weights corresponding to the fractional isotopic abundances [51]. Table I also includes the calculated nuclear sensitivity coefficients for both elements.

The δR_{Ir} correction to the assumed natural abundance-averaged nuclear charge radius $R_{\text{Ir},0}$ was calculated using both the MCDHF [42] and RMBPT [31] theoretical methods using Eq. (1):

$$\delta R_{\text{Ir}} = \frac{1}{S_{\text{Ir}}} \{ \delta E_{\text{Ir-Os}}^{\text{exp}} - [E_{\text{Ir}}^{\text{th}}(R_{\text{Ir},0}) - E_{\text{Os}}^{\text{th}}(R_{\text{Os}})] \}. \quad (2)$$

Applying the method to abundance-averaged values results in an abundance-averaged nuclear charge radius for Ir. This is possible because of the precise nuclear charge radii of the naturally abundant Os isotopes in the literature [19]. The nuclear charge radius of ^{191}Ir can be calculated from the abundance-averaged value, based on the well-known (i) relative abundances of the Ir isotopes [51] and (ii) differences between the mean squared charge radii of Ir isotopes from optical measurements by Verney *et al.* [27,36,37].

Uncertainty assessment. The uncertainty $\Delta(\delta R_{\text{Ir}})$ in the charge radius correction δR_{Ir} follows from Eq. (2):

$$\Delta(\delta R_{\text{Ir}}) = \left(\frac{\{ \Delta[E_{\text{Ir}}(R_{\text{Ir}}) - E_{\text{Os}}(R_{\text{Os}})] \}^2}{S_{\text{Ir}}^2} + (\delta R_{\text{Ir}})^2 \left(\frac{\Delta S_{\text{Ir}}}{S_{\text{Ir}}} \right)^2 + \frac{(\Delta E_{\text{Ir-Os}}^{\text{exp}})^2}{S_{\text{Ir}}^2} + \frac{(S_{\text{Os}} \Delta R_{\text{Os}})^2}{S_{\text{Ir}}^2} \right)^{1/2}. \quad (3)$$

It has four main contributions: (i) the uncertainty $\Delta[E_{\text{Ir}}(R_{\text{Ir},0}) - E_{\text{Os}}(R_{\text{Os}})]$ in the theoretical energy difference without the uncertainty associated with the nuclear charge radius of Os, (ii) the uncertainty in the nuclear sensitivity coefficient ΔS_{Ir} , (iii) the uncertainty in the measured energy difference $\Delta E_{\text{Ir-Os}}^{\text{exp}}$, and (iv) the uncertainty ΔR_{Os} in the natural abundance averaged charge radius of Os.

The main reason for the small overall nuclear charge radius uncertainty is the tight assessment of the uncertainties of the terms of Eq. (3). Uncertainty (i), for example, is overestimated if individual transition energy uncertainties of Ref. [31] are added in quadrature. This is because most of the theoretical uncertainty contributions are systematic and smooth functions of Z and cancel out in the theoretical energy difference.

In calculations of the absolute transition energy [31], uncertainties arise from omitted theory terms. Purely numerical errors in the calculation (e.g., number of grid points used, basis-set truncations) can be controlled to be negligible. The uncertainties due to omitted theory terms can be estimated from Z -dependent uncertainties given in Ref. [31] and represented as a smooth function $F_{\text{Theory}}(Z)$. Taking the uncertainty in the transition energy differences $\Delta[E_{\text{Ir}}(R_{\text{Ir}}) - E_{\text{Os}}(R_{\text{Os}})] = F_{\text{Theory}}(Z = 77) - F_{\text{Theory}}(Z = 76)$, the uncertainty of the difference between two absolute energies can be estimated to be more than an order of magnitude smaller than the individual transition energy uncertainties.

Detailed analysis of the contributing uncertainties, including effects of neglected mass terms, resulted in the uncertainty contributions listed in Table II. The theoretical energy difference calculated using RMBPT and MCDHF agree within the theoretical uncertainties estimated above.

For the experimental uncertainty term, the calibration uncertainty contributed significantly to the overall uncertainty in both methods of experimental determination. The uncertainty in the calibration dispersion was found through numerical integration of the dispersion function uncertainty across the determined separation. Investigations into Gaussian line shape bias for asymmetric line profiles were performed and found to have negligible effect.

The last term in Eq. (3) was determined using the uncertainty in the Os natural-abundance-averaged nuclear charge radius, with the values listed in Ref. [19].

TABLE II. Values are the variances of the quantities given in Eq. (3) using the measured absolute line center difference for first and second diffraction orders. The notation listed in the table is as follows: $O_s/I_{r, \text{exp, stat}}$ is the experimental statistical uncertainty, $O_s/I_{r, \text{exp, cal}}$ is the experimental calibration uncertainty, $O_{s, \text{ref}}$ is the uncertainty of the natural-abundance-averaged charge radius of Os nuclides, δE_{th} is the uncertainty in the difference of theoretically calculated energies, and S_{Ir} is the uncertainty in the nuclear sensitivity coefficient.

Uncertainty contribution	First order (10^{-6} fm ²)	Variance	Second order (10^{-6} fm ²)	Variance
$O_{s, \text{exp, stat}}$ (iii)	7.78	15.4%	15.7	14.8%
$I_{r, \text{exp, stat}}$ (iii)	4.77	9.4%	40.4	38.1%
$O_{s, \text{exp, cal}}$ (iii)	12.4	24.3%	19.2	18.1%
$I_{r, \text{exp, cal}}$ (iii)	12.8	25.3%	17.6	16.6%
$\text{Experiment}_{\text{total}}$	37.7	74.4%	92.9	87.7%
$O_{s, \text{ref}}$ (iv)	1.83	3.6%	1.83	1.8%
δE_{th} (i)	7.22	14.3%	7.22	6.8%
S_{Ir} (ii)	3.90	7.7%	3.90	3.7%
$\text{Theory}_{\text{total}}$	13.0	25.6%	13.0	12.3%
Total	50.7	100.0%	105.9	100.0%

Results and discussion. The experiments presented here allowed us to determine the natural abundance-averaged nuclear charge radius of iridium to be 5.439(12) fm, which is the average of the RMBPT and MCDHF results. With this and the OIS measurements by Verney *et al.* [27] along the Ir isotopic chain, our measurement reduces the uncertainties of the absolute values across all the isotopes included in the OIS experiment.

Combining our results with the Ir OIS measurement of $R_{193}^2 - R_{191}^2 = 0.0691(8)$ [27] leads to the nuclear radii of ^{191}Ir and ^{193}Ir being 5.435(12) fm and 5.441(12) fm, respectively. This determination is almost an order of magnitude more precise than the values in Ref. [19]. The uncertainty of our measurement is dominated by experimental uncertainties representing $>70\%$ of the total, which can potentially be improved in future measurements.

As an additional check of the validity of the method, we have compared data available in the literature for Pt, W, and Pb, all with well-known nuclear charge radii [19]. The $3s\ ^2S_{1/2} - 3p\ ^2P_{3/2}$ Na-like D2 x-ray transitions have been measured for these elements [31,52], allowing the energy differences to be calculated to 10^{-3} relative uncertainties.

By correcting the theoretical energy values of Ref. [31] with the nuclear charge radii from Ref. [19], the experimental difference results agree with the theory within their uncertainties. This evaluation provides us further confidence in this technique.

Charge radius measurements of isotopes in this region have been scarce over the years, leading to tabulated values in the literature [19,22] having large uncertainties for some isotopes. Measurements and analyses of muonic x-ray spectra of Hf, Ta, W, Os, and Ir isotopes were performed by De Wit *et al.* [53], who obtained nuclear parameters from which their charge radii were derived in Ref. [54]. In the following years, measurements of high precision by muonic atom spectroscopy and elastic electron scattering helped improve the accuracy of the charge radius of most of these isotopes. Recently, Ir OIS measurements have been conducted by Verney *et al.* [27], but since the measurements by De Wit *et al.* [53], the charge radii of Ir isotopes have not been remeasured with higher precision.

Conclusions. We have demonstrated a technique based on Na-like ions to determine nuclear charge radii. In this measurement, the nuclear charge radius of ^{191}Ir has been extracted, and the uncertainty from previously reported values has been improved by a factor of 8. Combining our results with precise OIS measurements [27] produces improved absolute Ir nuclear charge radii across the full isotopic sequence. Future work will include the utilization of other nearby charge states to further reduce the uncertainty of the value reported in this letter. This technique may prove useful in extracting precise nuclear charge radii of highly deformed nuclei that can be used for symmetry-breaking investigations [55]. The method can be applied to radioactive isotopes, particularly heavily deformed nuclei in the Rn to Pa region to achieve similar levels of uncertainty at rare isotope facilities such as TRIUMF [56].

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Data availability. The experimental Na-like D1 wavelength positions in first- and second-order diffraction were found through Gaussian fits of the respective spectral lines. Potential blended features were remedied with the technique described in Ref. [48] using time-adjacent Os and Ir spectra.

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Correction: Values given for the nuclear charge radius in the abstract and in the second paragraph of the “Results and discussion” section have been corrected.