Highly Charged Ions for Atomic Clocks, Quantum Information, and Search for α variation

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(Received 16 May 2014; published 16 July 2014)

We propose 10 highly charged ions as candidates for the development of next generation atomic clocks, quantum information, and search for α variation. They have long-lived metastable states with transition wavelengths to the ground state between 170–3000 nm, relatively simple electronic structure, stable isotopes, and high sensitivity to α variation (e.g., Sm^{14+}, Pr^{10+}, Sm^{13+}, Nd^{10+}). We predict their properties crucial for the experimental exploration and highlight particularly attractive systems for these applications.

DOI: 10.1103/PhysRevLett.113.030801 PACS numbers: 06.30.Ft, 06.20.Jr, 31.15.ac, 32.30.Jc

The past few years have marked unprecedented improvements in both the accuracy and stability of optical frequency standards [1–5]. The Sr lattice clock group has just reported the achievement of the 6.4×10^{-18} accuracy [3]. This remarkable progress poses the question of what are the novel schemes for the next generation of clock development that may achieve accuracy at the next decimal point, 10^{-19}. This work proposes 10 highly charged ions (HCI) as candidates for the development of next generation atomic clocks and other applications. Further development of even more precise frequency standards is essential for new tests of fundamental physics, search for the variation of fundamental constants, very-long-baseline interferometry for telescope array synchronization, and development of extremely sensitive quantum-based tools for geodesy, hydrology, and climate change studies, inertial navigation, and tracking of deep-space probes [2,3].

Modern theories directed toward unifying gravitation with the three other fundamental interactions suggest variation of the fundamental constants in an expanding Universe [6]. Studies of quasar absorption spectra indicate that the fine-structure constant, $\alpha = e^2/\hbar c$, may vary on a cosmological space-time scale [7,8]. This result has not been confirmed yet by other groups (see [9] and references therein). As a result, the status of the observational search for α variation is presently unclear and further investigations are required [10]. Spatial α-variation hypothesis can be tested in terrestrial studies if sensitivity $\delta\alpha/\alpha \sim 10^{-19}$ yr^{-1} is achieved [11]. The current best limit of $\delta\alpha/\alpha \sim 10^{-17}$ yr^{-1} comes from measuring the frequency ratio of Al^+ and Hg^+ clocks [12].

The signature feature of the system suitable for atomic clock development and subsequent tests of α variation is the availability of the optical (or near-optical) transition between a ground state and a long-lived metastable state. It appears at first glance that this requirement reduces atomic clocks based on HCI to transitions between states of the ground-state configuration (see, e.g., [13,14]) which are not sensitive to variation of α. However, this turns out not to be the case. Despite very large ionization energies, certain ions have transitions belonging to different configurations that lie in the optical range due to level crossings and strongly enhanced sensitivity to variation of α [15,16]. Highly charged ions are less sensitive to external perturbations than either neutral atoms or singly charged ions due to their more compact size. Recent studies of uncertainties [14,17] have shown that the fractional accuracy of the transition frequency in the clocks based on HCI can be smaller than 10^{-19}.

One of the main obstacles for experimental work toward the development of highly charged ions for precision measurement applications is the lack of experimental data for these HCI systems as well as difficulties in accurate theoretical predictions of the transition wavelengths owing to severe cancellations of energies of upper and lower states (since we are interested in working near level crossing). The goal of the present work is to resolve this problem. We identified all of the highly charged ions that are particularly well suited for experimental exploration, i.e., that satisfy the following criteria: (1) the existence of long-lived metastable states with transition frequencies to the ground states ranging between $(0.1–1.8) \times 10^{15}$ Hz, (2) high sensitivity to α variation, and (3) the existence of stable isotopes. Other practical considerations include relatively simple electronic structure, i.e., with one, two, or three valence electrons above the closed core, which excludes

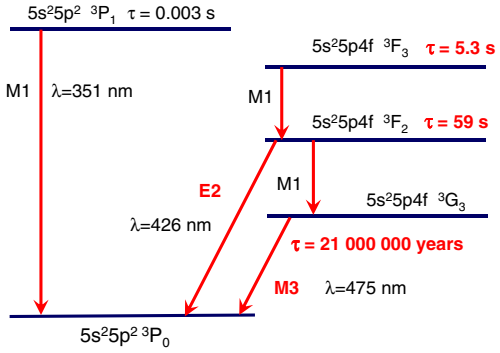

FIG. 1 (color online). Energy levels and radiative lifetimes of low-lying levels of Sn-like Pr^{9+}.

core-excited states such as those considered in [16]. We find that only the ions in four isoelectronic sequences, Ag-like, Cd-like, In-like, and Sn-like, satisfy these criteria. There are no useful level crossings for isoelectronic sequences corresponding to the previous row of the periodic table [18]. The level crossings for the isoelectronic sequences in the next row of the periodic table that includes Au-like, Hg-like, Tl-like, and Hg-like ions occur for ions with no stable isotopes [18].

Our study of the HCIs showed that some of these systems have several metastable states representing a level structure and other properties that are not present in any neutral atom and low-ionization state ions and may be advantageous not only for the development of atomic clocks but also for providing new possibilities for quantum information storage and processing. For example, we find that the first excited state in Sn-like Pr^{9+} can be counted as a second "ground" state with an estimated lifetime of 10^{14} seconds since the fastest transition contributing to the lifetime is $M3$. The low-lying levels of the Sn-like Pr^{9+} ion and our estimates of the radiative lifetimes are shown in Fig. 1 for illustration. A much more compact size of the HCI electronic cloud may significantly reduce decoherence processes due to an undesired external perturbation, and extremely long-lived states may provide a resource for quantum memories. The large charge may allow for a faster quantum logic gate operation [19].

While highly charged ions lack strong electric-dipole transitions for laser cooling, sympathetic cooling may be employed similar to the case of the Al^+ clock, which is cooled using a laser-cooled Be^+ or Mg^+ ion [1]. Experimental investigations toward the sympathetic cooling of HCIs and the precision laser spectroscopy of forbidden transitions are currently in progress [20–23]. Recently, the evaporative cooling of Ar^{16+} in a Penning trap was demonstrated [20]. Storage and cooling of highly charged ions require ultrahigh vacuum levels that can be obtained by cryogenic methods. A linear Paul trap operating at 4 K capable of very long ion storage times of about 30 h was recently developed in [22].

Methods.—We use two different relativistic high-precision all-order approaches for all of the calculations carried out in this work. The first approach is the relativistic linearized coupled-cluster method that includes all single, double, and partial triple excitations of Dirac-Fock wave function [24]. It has been extremely successful in predicting the properties of alkali-metal atoms and monovalent ions [24]. The second approach combines the modified linearized coupled-cluster method with the configuration interaction (CI) [25,26]. This CI+all-order method yielded accurate atomic properties for a number of divalent systems and trivalent Tl [26–32]. This work presents the first application of the CI+all-order method for four-valence-electron systems such as Sn-like ions. We demonstrated that the CI+all-order method can accurately predict properties of systems with four valence electrons (using test systems where the experimental results are known) and

TABLE I. Comparison of the energies of Ag-like Nd^{13+}, Sm^{15+}, and In-like Ce^{9+} ions relative to the ground state with the experiment [35,36] and other theory [17]. Differences with the experiment are given in cm^{-1} and % in columns "Diff."

Ion	Level	Ref. [17]	Experiment	Present	Differences	Differences %
Nd^{13+}	$5s_{1/2}$	0	0	0	0	
	$4f_{5/2}$	58 897	55 870	55 706	164	0.29%
	$4f_{7/2}$	63 613	60 300	60 134	166	0.28%
	$5p_{1/2}$		185 066	185 028	38	0.02%
	$5p_{3/2}$		234 864	234 887	−23	−0.01%
Sm^{15+}	$4f_{5/2}$	0	0	0	0	
	$4f_{7/2}$	6806	6555	6444	111	1.69%
	$5s_{1/2}$	55 675	60 384	60 517	−133	−0.22%
	$5p_{1/2}$		268 488	268 604	−116	−0.04%
	$5p_{3/2}$		333 203	333 385	−182	−0.05%
Ce^{9+}	$5p_{1/2}$		0	0	0	
	$5p_{3/2}$		33 427	33 450	−23	−0.07%
	$4f_{5/2}$		54 947	54 683	264	0.48%
	$4f_{7/2}$		57 520	57 235	285	0.50%

TABLE II. Energies and α-variation sensitivity coefficients q for Ag-like and In-like ions relative to the ground state in cm^{-1}; $K = 2q/\omega$ is the enhancement factor. Wavelengths λ (in nm) for transitions to the ground states and total radiative lifetimes τ are listed. Experimental values are given for the wavelengths [35,36] in Nd^{13+}, Sm^{15+}, and Ce^{9+}.

Level	Energy	q	K	λ	τ
Ag-like Nd^{13+}	$5s_{1/2}$ ground state				
$4f_{5/2}$	55 706	104 229	3.7	179.0	1.3×10^6 s
$4f_{7/2}$	60 134	108 243	3.6	165.8	0.996 s
$5p_{1/2}$	185 028	15 953	0.2	54.03	0.204 ns
$5p_{3/2}$	234 887	72 079	0.6	42.58	0.098 ns
Ag-like Sm^{15+}	$4f_{5/2}$ ground state				
$4f_{7/2}$	6444	5910	1.8	1526	0.308 s
$5s_{1/2}$	60 517	−134148	−4.4	165.6	3.1×10^5 s
$5p_{1/2}$	268 604	−114999	−0.9	37.25	0.167 ns
$5p_{3/2}$	333 385	−41477	−0.2	30.01	0.0731 ns
In-like Ce^{9+}	$5p_{1/2}$ ground state				
$5p_{3/2}$	33 450	37 544	2.2	299.2	0.0030 s
$4f_{5/2}$	54 683	62 873	2.3	182.0	0.0812 s
$4f_{7/2}$	57 235	65 150	2.3	173.9	2.18 s
In-like Pr^{10+}	$5p_{1/2}$ ground state				
$4f_{5/2}$	3702	73 849	40	2700(140)	8.5×10^4 s
$4f_{7/2}$	7031	76 833	22	1422(40)	2.35 s
$5p_{3/2}$	39 141	44 098	2.3	255.5(3)	0.0018 s
In-like Nd^{11+}	$4f_{5/2}$ ground state				
$4f_{7/2}$	4180	3785	1.8	2392(60)	1.19 s
$5p_{1/2}$	53 684	−85692	−3.2	186.3(1.7)	0.061 s
$5p_{3/2}$	99 066	−34349	−0.7	100.9(5)	0.000 88 s

designed efficient algorithms to construct four-electron CI configuration spaces that contain all important sets of configurations.

An accurate calculation of the highly charged ion energies and corresponding wavelengths not only requires the all-order methods to treat the Coulomb correlation effects, but also an accurate treatment of the Breit interaction, QED corrections, and inclusion of the high partial wave contribution. We treat the Breit interaction on the same footing as the Coulomb interaction in the basis set. The QED radiative corrections to the energy levels were included using the method described in [33]. The partial waves with $l_{\max} = 6$ are included in all summations in many-body perturbation theory or all-order terms, and $l > 6$ contributions are extrapolated. We find that inclusion of the higher partial waves with $l > 6$ is very important for an accurate description of the $4f$ states. We illustrate the size of various contributions to the energies of Ag-like Nd^{13+}, In-like Pr^{10+}, and Sn-like Pr^{9+} ions in Table I of the Supplemental Material [34] to show that all of these contributions are essential for accurate determination of the positions of the energy levels and transition wavelengths of the HCIs.

To further verify the accuracy of our calculations, we compared our values for the three ions where reliable spectra identification provides the experimental values of the energy levels. Comparison of the energies of Ag-like Nd^{13+}, and Sm^{15+} ions and In-like Ce^{9+} relative to the ground state with experiment [35] and other theory [17] is given in Table I. Our values are in excellent agreement with the experiment for all three cases.

Results.—In our study, we considered the following most essential properties for experimental development: ground-state ionization energies, energy levels, transition wavelengths, lifetimes, and sensitivity to the variation of the fine-structure constant α. The sensitivity of the atomic transition frequency ω to the variation of α can be quantified using a coefficient q defined as $\omega(x) = \omega_0 + qx$, where $x = (\alpha/\alpha_0)^2 - 1$ and the frequency ω_0 corresponds to the value of the fine-structure constant α_0 at some initial point in time. In the experiment, the ratio of two frequencies, which is a dimensionless quantity, is studied over time. Therefore, it is preferable to select transitions with significantly different values of q. It is convenient to define the dimensionless enhancement factor $K = 2q/\omega$ in order to compare the sensitivity to α between the transitions with significantly different frequencies as a wide range of transition frequencies is valuable in HCIs. To calculate the value of the sensitivity coefficient q, we carry out three calculations with different values of α for every ion considered in this work. In the first calculation, the current CODATA value of $\alpha = 7.2973525698(24) \times 10^{-3}$ [37] is used. In the other two calculations, the value of α^2 is varied by $\pm 1\%$. The value of q is then determined as a numerical derivative.

The results for Ag-like ions and In-like ions that can be considered "monovalent," i.e., with one valence electron

TABLE III. Energies and α-variation sensitivity coefficients q for Cd-like, In-like, and Sn-like ions relative to the ground state in cm^{-1}; $K = 2q/\omega$ is the enhancement factor. Wavelengths λ (in nm) for transitions to the ground states and total radiative lifetimes τ are listed.

Level	Energy	q	K	λ	τ
Cd-like Nd^{12+}	$5s^2\,{}^1S_0$ ground state				
$5s4f\,{}^3F_2$	79 469	101 461	2.6	125.8(9)	8.5×10^{10} s
$5s4f\,{}^3F_3$	80 769	102 325	2.5	123.8(9)	19.7 s
$5s4f\,{}^3F_4$	83 730	105 340	2.5	119.4(9)	1.95 s
$5s5p\,{}^3P_1$	168 547	19 465	0.2	59.3(3)	1.80 ns
Cd-like Sm^{14+}	$4f^2\,{}^3H_4$ ground state				
$5s4f\,{}^3F_2$	2172	−127720	−118	4600	5.6×10^{13} s
$5s4f\,{}^3F_3$	3826	−126746	−66	2614(470)	8.515 s
$4f^2\,{}^3H_5$	4939	4917	2.0	2025(40)	0.313 s
$5s4f\,{}^3F_4$	8463	−121952	−29	1182(100)	0.556 s
In-like Sm^{13+}	$5s^24f_{5/2}\,{}^2F_{5/2}$ ground state				
$5s^24f\,{}^2F_{7/2}$	6203	5654	1.8	1612(28)	0.367 s
$4f^25s\,{}^4H_{7/2}$	20 254	123 621	12	494(22)	0.133 s
$4f^25s\,{}^4H_{9/2}$	22 519	125 397	11	444(18)	0.141 s
$4f^25s\,{}^4H_{11/2}$	25 904	128 875	10	386(14)	0.576 s
Sn-like Pr^{9+}	$5p^2\,{}^3P_0$ ground state				
$5p4f\,{}^3G_3$	20 216	42 721	4.2	475(18)	6.6×10^{14} s
$5p4f\,{}^3F_2$	22 772	42 865	3.8	426(13)	59.0 s
$5p4f\,{}^3F_3$	25 362	47 076	3.7	382(12)	5.33 s
$5p4f\,{}^3F_4$	27 536	37 197	2.7	352(11)	0.234 s
$5p^2\,{}^3P_1$	28 512	47 483	3.3	351.2(5)	0.002 96 s
Sn-like Nd^{10+}	$4f^2 J = 4$ ground state				
$5p4f\,J = 3$	1564	−81052	−104		1.6×10^4 s
$4f^2\,J = 5$	3059	3113	2.0		1.4 s
$5p4f\,J = 2$	5060	−60350	−24	2200(430)	25 s
$4f^2\,J = 6$	6222	5930	1.9	1610(110)	1.4 s
$5p4f\,J = 3$	7095	−63285	−18	1480(240)	3.9 s

above the closed $5s^2$ shell, are given in Table II. Energies and α-variation sensitivity coefficients q relative to the ground state in cm^{-1}, enhancement factor $K = 2q/\omega$, wavelengths (in nm) for transitions to the ground states, and total radiative lifetimes τ are listed. Experimental values are given for the wavelengths [35,36] of the first three ions. We developed several methods to evaluate uncertainties in the values of the theoretical wavelengths based on the size of all four corrections described above and comparison with experiment for previous ions of the corresponding isoelectronic sequences, for example, Ba^{6+}–Ba^{9+}. The uncertainties for the wavelength transitions to the ground states are listed in parenthesis for Pr^{10+} and Nd^{11+}.

The $5s$-$4f$ level crossing in an Ag-like isoelectronic sequence happens from Nd^{13+} to Sm^{15+}. The Pm^{14+} has no stable isotopes and the $5s$ and $4f_{5/2}$ states in Pm^{14+} are separated by only about 300 cm^{-1}. Nd^{13+} represents a particularly attractive case since the strongest transition from the metastable $4f_{5/2}$ level of this ion is $E3$, resulting in the extremely long lifetime of more than 15 days. Therefore, this system may be considered to have two ground states. While Nd^{13+} transition $5s$-$4f$ wavelength is in UV, another ion can be used to probe the transitions as in the Al^+ clock scheme which also has a clock transition in UV. Many of the low-lying transitions in all ions considered here have large q and K coefficients indicating large sensitivities to α variation. Our calculations of K for infrared transitions in Pr^{10+} give $K = 22$ and $K = 40$. For comparison, $K = 3$ [38] for UV transition in Hg^+ which provided the most precise current test of α variation in Al^+ and Hg^+ clocks carried out in [12]. The Al^+ ion is light and does not produce additional enhancement of sensitivity to α variation.

There are two level crossings of interest for the present work in an In-like isoelectronic sequence, $5p$-$4f$ and $4f$-$5s$. The first one happens for Pr^{10+} and Nd^{11+} and leads to a change of level order from $5p$, $4f$ to $4f$, $5p$. Pr^{10+} represents a particularly attractive case where both $4f_j$ levels are located between the $5p_{1/2}$ and $5p_{3/2}$ fine structure multiplet, making $4f_{5/2}$ a very long-lived metastable level with 1 day radiative lifetime. The order of levels changes again for Sm^{13+}, where the $5s4f^2$ configuration becomes the closest to the ground $5s^24f_j$ fine-structure multiplet. This leads to a very interesting level structure with a metastable $5s4f^2$ $J = 7/2$ level in the optical transition range to both ground and excited $5s^24f_{7/2}$ levels of the fine-structure multiplet. The results for this In-like ion are listed in Table III together with the results of other HCIs in Cd-like and Sn-like isoelectronic sequences.

TABLE IV. Ionization energies (IE) of Ag-like and In-like ions in cm^{-1} and eV. Comparison with experiment [39] is given where available. Ground-state configurations of the valence electron are listed.

Ion		Level	IE (cm^{-1})	IE (eV)	Experiment (eV)
Ag-like	Ba^{9+}	$5s$	1 181 600	146.5	146.52(12)
Ag-like	Nd^{13+}	$5s$	1 960 539	243.1	
Ag-like	Sm^{15+}	$4f_{5/2}$	2 471 848	306.5	
In-like	Ca^{6+}	$5p_{1/2}$	671 692	83.3	82.9(2.0)
In-like	Ba^{7+}	$5p_{1/2}$	816 588	101.2	101.0(2.1)
In-like	Ce^{9+}	$5p_{1/2}$	1 138 158	141.1	
In-like	Pr^{10+}	$5p_{1/2}$	1 314 931	163.0	
In-like	Nd^{11+}	$4f_{5/2}$	1 554 148	192.7	

The $5s$-$4f$ level crossing in Cd-like ions happens for Nd^{12+}-Sm^{14+} ions. The strongest transition from the first excited levels of both ions is $M2$, resulting in extremely long lifetimes. The case of Nd^{12+} is very similar to Ag-like Nd^{13+} but the wavelengths are further in UV. We note that q values are positive for Nd^{12+} and negative for Sm^{14+} which creates additional enhancements for an α-variation search if the relative transition frequencies in Nd^{12+} and Sm^{14+} are monitored. Similar cases are found in other isoelectronic sequences studied here because the energy levels change order with the level crossings and the values of q in ions before and after the crossing have an opposite sign.

The ions of interest in the Sn-like isoelectronic sequence are Pr^{9+} and Nd^{10+}. The case of Pr^{9+} is particularly interesting, since the lowest metastable state $5p4f\ J = 3$ is extremely long lived, with transition to the ground state being in the optical range, 495 nm (see Fig. 1). The strongest allowed transition is $M3$ making this ion a unique system. The next two levels also have optical transitions to the ground state and are metastable with 59 and 5.3 s lifetimes. Moreover, there is a relatively strong $M1$ transition to the ground state from $5p^2\ ^3P_1$ at 351 nm that may be potentially used for cooling and probing. The ground and first excited states of Nd^{10+} are extremely close and the resulting uncertainty is on the order of the transition energy. While our calculations place $4f^2$ to be the ground state, the higher-order corrections are particularly large in this case, almost 3 times that of the transition energy, which might lead to the placement of the $5p4f\ J = 3$ as the ground state.

The ionization potentials for Ag-like and In-like ions are given in Table IV. We note that their values mainly depend on the degree of ionization (147 eV for Ag-like Ba^{9+} and 141 eV for In-like Ce^{9+}) so this table may be used to estimate the ionization potentials for Cd-like and Sn-like ions as well.

In summary, we provided a clear direction for the further experimental study of highly charged ions for the development of next-generation atomic clocks and search for α variation. We coducted a first high-precision study of such systems as well as demonstrated the first treatment of any systems with four valence electrons (Sn-like ions) using the all-order methodology. We identified *all* ions that fit the criteria for the most straightforward experimental exploration including sensitivity to α variation. We also proposed the use of highly charged ions for quantum information research. The ions listed in this work present a completely unexplored resource for quantum information due to their unique atomic properties and potential reduced sensitivity to decoherence effects. We hope that this work will lay the foundation for the future development of this field.

We thank C. W. Clark, C. Monroe, J. Tan, Yu. Ralchenko, and P. Beiersdorfer for useful discussions. This work was supported in part by U.S. NSF Grant No. PHY-1212442. M. G. K. acknowledges support from RFBR Grant No. 14-02-00241. M. S. S. thanks the School of Physics at UNSW, Sydney, Australia for hospitality and acknowledges support from the Gordon Godfrey Fellowship program, UNSW. The work is partly supported by the Australian Research Council.

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