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Rapid Communications

Selected highly charged ions as prospective candidates for optical clocks with quality factors larger than 10^{15}

Yan-mei Yu[1,*] and B. K. Sahoo[2,†]
[1]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*
[2]*Atomic, Molecular and Optical Physics Division, Physical Research Laboratory, Navrangpura, Ahmedabad 380009, India*
and State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China

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The Ni^{12+}, Cu^{13+}, Pd^{12+}, and Ag^{13+} highly charged ions (HCIs) are proposed for making very accurate optical clocks with the fractional uncertainties below 10^{-19} level. These HCIs have simple atomic energy levels, clock transitions with quality factors larger than 10^{15}, and optical magnetic-dipole ($M1$) transitions that can be used for laser cooling and detecting quantum jumps on the clock transitions by the shelving method. To demonstrate the projected fractional uncertainties, we estimate orders of magnitude of the Zeeman, Stark, blackbody radiation, and electric quadrupole shifts of the clock transitions by performing calculations of the relevant atomic properties in the above HCIs.

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Investigation of spectral properties of highly charged ions (HCIs) has a very long history. Their spectra are used for identifying an abundance of elements in the solar corona and other astrophysical objects, describing exotic phenomena in the nuclear reactions, diagnosing plasma processes, etc. [1–4]. However, only recently are HCIs recognized as potential candidates for making ultraprecise atomic clocks, developing tools for quantum information, and probing possible variation of the fine-structure constant [5–10]. It is generally intended to use forbidden transitions in atomic clocks as they provide narrow linewidths. Choosing the forbidden transitions from the HCIs is more advantageous for atomic clocks compared with those of neural atoms and singly charged ions, because the HCIs are less sensitive to external perturbations owing to their strongly contracted atomic orbitals. However, most of the transition frequencies in HCIs are in the x-ray region and beyond the reachable range of the narrow-bandwidth lasers in laboratories. Only a very few HCIs, belonging to two specific categories, have been suggested for making optical clocks. One of the possibilities is to choose the transitions at the level crossings of two nearly degenerate states [6–9], and the other is to consider the magnetic dipole ($M1$) transitions among the fine-structure and hyperfine-structure manifolds [5,10]. Some of the previously proposed HCIs for atomic clocks have very complicated atomic structures to carry out experiments. It is also strenuous to perform high-precision calculations in those ions for theoretical studies. The suggested $M1$ clock transitions in these HCIs have short lifetimes, which corresponds to small quality factors (Q). Again, most of the earlier proposed HCI clock candidates lack appropriate optical cooling transitions. Laser trapping and cooling of the clock candidates are immensely beneficial, which can enhance the interrogation time and reduce the Doppler shifts sharply for the ultrahigh-precision measurements. Therefore, it is desirable to find HCIs that can provide both clock and cooling transitions in the optical region.

In this Rapid Communication, we propose a class of HCIs for optical atomic clocks aiming at the 10^{19} precision level. Particularly, the S-like Ni^{12+} and Cu^{13+} ions having the $3p^4$ ground-state configuration and the Se-like Pd^{12+} and Ag^{13+} ions having the $4p^4$ ground-state configuration possess unique features that are suitable for the optical frequency standards. We outline some of the important features of these clock candidates here: (i) The $E2$ transitions between the $^3P_2 - {}^3P_0$ intercombination levels of the ground states of these ions, as shown in Fig. 1, are found to be in the optical region with Q values ranging between 10^{15} and 10^{16}. On the other hand, these transitions are not so highly forbidden like the electric octupole ($E3$) clock transition of the $^{171}\mathrm{Yb}^+$ ion [11], hence they can be directly interrogated within a stipulated time period. (ii) The intercombination lines offer several strong $M1$ transitions in the optical region that can be used for the ion cooling, state manipulation, and detection techniques. (iii) We demonstrate later that most of the typical systematic effects of an atomic clock experiment, such as Zeeman, Stark, blackbody radiation (BBR), and electric quadrupole shifts, are estimated to be negligibly small in these ions. This projects out the maximum fractional uncertainties to the clock frequencies (ν) to be below 10^{-19} level in these HCIs. (iv) The atomic level structures of the undertaken ions are comparatively simpler than the earlier proposed HCIs for the optical clocks. Two isotopes have nuclear spin $I = 0$, which leads to suppression of the second-order Zeeman effect. (v) Last but not least, to mention that the Ni and Cu HCIs are abundant in the astrophysical objects [3]; thus one can make a comparative analysis between the astrophysical observations with the clock measurements to infer about possible temporal variation of the fine-structure constant [4].

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[*] ymyu@aphy.iphy.ac.cn
[†] bijaya@prl.res.in

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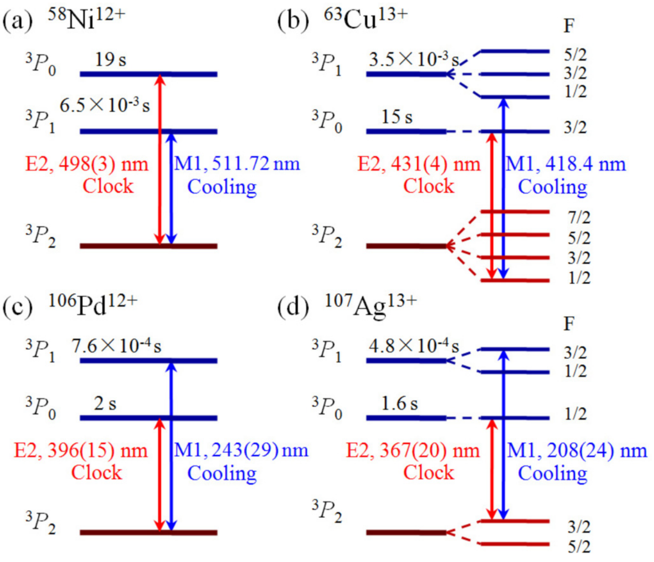

FIG. 1. Schematic diagrams of the low-lying atomic and hyperfine energy levels of the (a) ^{58}Ni^{12+}, (b) ^{63}Cu^{13+}, (c) ^{106}Pd^{12+}, and (d) ^{107}Ag^{13+} HCIs. Intercombination lines that are apt for the clock and cooling transitions are shown in red and blue, respectively. The estimated wavelengths and lifetimes of the excited states in the clock and cooling transitions are mentioned.

TABLE I. The estimated values of EEs, lifetimes (τ), wavelengths (λ), and transition rates (A_{ki}^{O}) due to the forbidden channels ($O = M1$ or $E2$) of the excited states to the ground states of the considered HCIs. Energies are compared with the available NIST data [20]. Other contributing transition rates to the lifetimes can be found in the Supplemental Material. Uncertainties are quoted within the parentheses. Numbers within the square brackets represent powers of 10.

	EE (in cm^{-1})		λ		A_{ki}^{O}	τ
Level	This work	NIST [20]	(nm)	O	(s^{-1})	(s)
	Ni^{12+} ion ($3p^4$ configuration)					
$^3P_1$	19560(20)	19541.8	511.72	$M1$	154	6.5[−3]
$^3P_0$	20251(450)	20060(100)	498(3)	$E2$	0.03	19
$^1D_2$	47512(2120)	47032.9	212.61	$M1$	261	3.6[−3]
$^1S_0$	99377(5130)	97836.2	102.21	$E2$	3.37	3.8[−4]
	Cu^{13+} ion ($3p^4$ configuration)					
$^3P_0$	23457(350)	23192(100)	431(4)	$E2$	0.06	15
$^3P_1$	23922(260)	23897	418.4	$M1$	283	3.5[−3]
$^1D_2$	53014(1560)	52540	190.3	$M1$	421	2.2[−3]
$^1S_0$	109356(4020)	107902	92.7	$E2$	4.34	2.4[−4]
	Pd^{12+} ion ($4p^4$ configuration)					
$^3P_0$	25275(470)		396(15)	$E2$	0.5	2
$^3P_1$	41190(2470)		243(29)	$M1$	1260	7.6[−4]
$^1D_2$	60037(500)		167(3)	$M1$	1155	8.5[−4]
$^1S_0$	119562(2390)		83.6(3)	$E2$	17.1	1.2[−4
	Ag^{13+} ion ($4p^4$ configuration)					
$^3P_0$	27242(760)		367(20)	$E2$	0.6	1.6
$^3P_1$	48028(2760)		208(24)	$M1$	1977	4.8[−4]
$^1D_2$	67514(540)		148(2)	$M1$	1736	5.6[−4]
$^1S_0$	134012(2540)		74.6(3)	$E2$	21.9	8.2[−5]

The basic spectroscopies of the aforementioned S-like HCIs have been investigated before for their extensive usage in the identification and interpretation of emission lines in astrophysics, fusion reactors, plasma diagnostics, etc. [12–16]. Most of these studies are concentrated on the allowed transitions, since their optical signals can be observed easily. The forbidden transitions between the $^3P_0$ and $^3P_2$ states, which are also important from the astrophysics point of view, have received less attention. On the other hand, the spectroscopies of the considered Se-like ions are not well known. We have employed the multiconfiguration Dirac-Hartree-Fock (MCDF) method of the GRASP2K program [17] and the Fock-space based relativistic coupled-cluster (FSCC) method of the DIRAC package [18] to carry out many spectroscopic properties of the above HCIs. Brief descriptions of both the methods and the calculated results of the excitation energies (EEs), the electric quadrupole moments, the electric dipole polarizabilities, and the transition probabilities of all the considered ions are given in the Supplemental Material [19] to support the following discussions.

Using the results from the MCDF method, we present the excitation energies and lifetimes of the excited states in Table I that are useful for the clock and cooling transitions of the investigated ions. The energies are compared with the available experimental values quoted in the National Institute of Science and Technology (NIST) database [20]. We have considered the Dirac-Coulomb Hamiltonian along with the Breit interaction and the lower-order QED effects in these calculations. The calculated energies in the Ni^{12+} and Cu^{13+} ions are in good agreement with the experimental values [20], from which the wavelengths of the proposed $^3P_0$-$^3P_2$ clock transitions are estimated to be about 2% accuracy. On this basis, we assume that our calculations for the Pd^{12+} and Ag^{13+} ions are of similar accuracies. We have also determined EEs of these ions by employing the FSCC method and find that these calculations agree well with the MCDF results (see the Supplemental Material). From the above table, the lifetime of the $^3P_2$ state is found to be more than 10 s in the Ni^{12+} and Cu^{13+} ions and about a few seconds in the Pd^{12+} and Ag^{13+} ions. From this analysis, we have chosen the $^3P_2$-$^3P_0$ transition in ^{58}Ni^{12+} and ^{106}Pd^{12+} with $I = 0$, the $|^3P_2, F = 1/2\rangle$-$|^3P_0, F = 3/2\rangle$ transition in ^{63}Cu^{13+} with $I = 3/2$, and the $|^3P_2, F = 3/2\rangle$-$|^3P_0, F = 1/2\rangle$ transition in ^{107}Ag^{13+} with $I = 1/2$ as the possible optical clock transitions. In the above table, we also give EEs and the lifetimes of the $^3P_1$, $^1D_2$, and $^1S_0$ states. The $M1$ transitions between the $^3P_1$-$^3P_2$ states are also in the optical region, which can be used for cooling and state manipulation techniques. The advantage of this $M1$ cooling transition is that it can offer an efficient method for detecting quantum jumps on the clock transitions by the usual electron shelving method rather than considering quantum logic spectroscopy.

It has been one of the most demanding tasks so far to find out suitable cooling transitions for the HCI atomic clocks. We have come up with a few stepwise strategies of cooling mechanism to cool down the proposed HCIs to the desired low temperature for the clock frequency measurements. As well known HCIs can be produced using one of the procedures such as accelerator, high-power laser, electron cyclotron resonance, electron

TABLE II. Values of ν, Q, Θ (in a.u.), α^{E1} (in a.u.), α^{M1} (in a.u.), A_{hf} (in MHz), and B_{hf} (in MHz) are listed along with the fractional systematic shifts for the respective HCIs. We have used nuclear magnetic dipole moment (μ_I) and electric quadrupole moment (Q_I) for ^{63}Cu as $\mu_I = 2.223\mu_N$ and $Q_I = -0.211b$, and for ^{107}Ag as $\mu_I = -1.135\mu_N$ and $Q_I = 0.98b$ in the determination of the A_{hf} and B_{hf} values.

Items	^{58}Ni^{12+}	^{106}Pd^{12+}	^{63}Cu^{13+}	^{107}Ag^{13+}
I	0	0	3/2	1/2
υ	6.01[14]	7.64[14]	6.95[14]	8.06[14]
Q	1.1[16]	1.5[15]	1.0[16]	1.3[15]
$A_{\mathrm{hf}}(^3P_2)$			10564	−1594
$B_{\mathrm{hf}}(^3P_2)$			−204	0
$\Theta(^3P_2)$	0.1465	0.1670	0.1260	0.1382
$\alpha^{E1}(^3P_2)$	0.2736	0.6531	0.2295	0.5617
$\alpha^{E1}(^3P_0)$	0.2751	0.6548	0.2306	0.5625
$\alpha^{M1}(^3P_2)$	2.10[a]	1.35[a]	−4.62[5][b]	−3.71[5][c]
$\alpha^{M1}(^3P_0)$	−149.85	4.66	103.76	−3.58
$\delta E^{(2)}_{\mathrm{Zeem}}/\upsilon$	3.77[−23]	−6.60[−25]	9.89[−20]	−6.85[−20]
$\delta E_{\mathrm{Stark}}/\upsilon$	−1.52[−24]	−1.43[−24]	−9.93[−25]	−7.33[−25]
$\delta E^{E1}_{\mathrm{BBR}}/\upsilon$	−2.10[−20]	−1.67[−20]	−1.37[−20]	−1.01[−20]
$\delta E^{M1}_{\mathrm{BBR}}/\upsilon$	−2.50[−20]	−3.85[−21]	7.88[−20]	1.60[−20]

[a] α^{M1} value $J = 2$.

[b] α^{M1} value $F = 1/2$.

[c] α^{M1} value $F = 3/2$.

beam ion sources, etc., techniques. Initially these ions can be trapped at very high temperature, in the order of megakelvin; then they can be decelerated by the traditional approach to reach a temperature around 10^5 K. At this stage, one may adopt the evaporation technique to bring the temperature down to 10^4 K with the storage time up to 1000 s [21]. Following this, a sympathetic cooling technique can be applied to decrease the temperature down to millikelvin as has been demonstrated in Ref. [22]. In this process the considered HCIs can be trapped simultaneously with another atomic species having a matched mass to charge ratio. In our case, the mass-to-charge ratios of the Pd^{12+} and Ag^{13+} ions match well with that of the Be$^+$ ion. Subsequently, the narrow $^3P_1$-$^3P_2$ lines in the Cu^{13+}, Pd^{12+}, and Ag^{13+} ions, as shown in Fig. 1, can be used for the laser cooling to achieve temperatures about nanokelvin. We analyze here the most commonly systematic effects in the atomic clock experiments for the considered HCIs. We list many properties of the atomic states associated with the clock transitions including I values of the relevant isotopes in Table II and give typical orders of magnitude of the systematic effects using these quantities. It is to be noted that these are the absolute values, but more precise values of the clock frequencies can be obtained by determining their uncertainties when the actual measurements are carried out. The Q values are estimated from the calculated lifetimes of the excited states given in Table I. We have evaluated the electric dipole polarizabilities (α^{E1}) and the electric quadrupole moments (Θ) by adopting the finite-field approach in the FSCC method [18] and the magnetic dipole (A_{hf}) and the electric quadrupole (B_{hf}) hyperfine-structure constants using the MCDF method [17]. More details of these results can be found in the Supplemental Material. We also give the magnetic dipole polarizabilities (α^{M1}) that may contribute significantly to the proposed clock transitions.

The first-order Zeeman shift is defined by $\delta E^{(1)}_{\mathrm{Zeem}} = \delta g \delta M \mu_B B$ for the external magnetic field B, Bohr magneton μ_B, and differential values of Lande g factors and M quantum numbers, δg and δM, respectively, of the clock states. The first-order Zeeman shift can be eliminated by choosing the $M_J = 0 \rightarrow M_J = 0$ transition in the Ni^{12+} and Pd^{12+} ions or measuring all the M components in the Cu^{13+} and Ag^{13+} ions. However, the second-order Zeeman shift, $\delta E^{(2)}_{\mathrm{Zeem}} = -\frac{1}{2}\delta\alpha^{M1}B^2$, can be significant, wherein $\delta\alpha^{M1}$ is the differential value of α^{M1} between the clock states. By defining the $M1$ operator $O^{M1} = (\vec{L} + 2\vec{S})\mu_B$ with $\vec{L}$ and $\vec{S}$ as the orbital and spin angular momentum operators, respectively, we evaluate α^{M1} for the $|\gamma J M_J\rangle$ state as

$$\alpha^{M1}(J) = -\frac{2}{3(2J+1)} \sum_{J'} \frac{|\langle J||O^{M1}||J'\rangle|^2}{E_J - E_{J'}}, \tag{1}$$

with the energies E and the reduced matrix element

$$\langle J||O^{M1}||J'\rangle = \mu_B \sqrt{S(S+1)(2S+1)(2J+1)} \times \sqrt{(2J'+1)} \begin{Bmatrix} L & S & J \\ 1 & J' & S \end{Bmatrix}. \tag{2}$$

For the hyperfine level $|F, M_F\rangle$, we estimate α^{M1} by considering only the dominant contributions from the hyperfine manifolds of the same principal (n) and angular momentum (J) states and neglecting contributions from other angular momentum states as

$$\alpha^{M1}(nIJF) = -\frac{2}{3(2F+1)} \sum_{F'} \frac{|\langle nIJF||O^{M1}||nIJF'\rangle|^2}{E_{nIJF} - E_{nIJF'}}, \tag{3}$$

in which the reduced matrix element of O^{M1} is given by

$$\langle nIJF||O^{M1}||nIJF'\rangle = \mu_B \sqrt{J(J+1)(2J+1)(2F+1)} \times \sqrt{(2F'+1)} \begin{Bmatrix} I & J & F \\ 1 & F' & J \end{Bmatrix} g_J. \tag{4}$$

Here, g_J is evaluated by

$$g_J = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}. \tag{5}$$

We have determined the hyperfine energy levels as

$$E_{nJF} = \frac{1}{2} A_{\mathrm{hf}} K + B_{\mathrm{hf}} \frac{(3/2)K(K+1) - 2I(I+1)J(J+1)}{4I(2I-1)J(2J-1)}, \tag{6}$$

where $K = F(F+1) - I(I+1) - J(J+1)$. Assuming a typical value of $B = 5 \times 10^{-8}$ T [6], $\delta E^{(2)}_{\mathrm{Zeem}}/\nu$ is found to be below 10^{-19}. However, we find that the fractional second-order Zeeman shifts in the ^{63}Cu^{13+} and ^{107}Ag^{13+} ions are close to the 10^{-19} level owing to small energy differences between the hyperfine levels. To suppress this systematic effect in these two ions, it may be needed to calibrate the magnetic field more stringently.

The α^{E1} value of a HCI scales as $1/Z_i^4$, where Z_i is the residual nuclear charge. Our calculation gives differential scalar polarizability values between the states associated with the clock transitions of Ni^{12+}, Cu^{13+}, Pd^{12+}, and Ag^{13+} as $\delta\alpha_0^{E1} \approx 10^{-3}$ a.u. These tiny $\delta\alpha_0^{E1}$ values lead to negligibly small fractional differential Stark shifts, $\delta E_{\mathrm{Stark}} = -\delta\alpha_0^{E1}\mathcal{E}^2/2$, to the clock transitions with the typical electric field strength $\mathcal{E} = 10$ V/m. Also, the light shifts caused by the probing and cooling lasers can be strongly suppressed by the very small $\delta\alpha_0^{E1}$ values. Here we have assumed that contribution due to the tensor polarizability of the $J = 2$ state, which is one order smaller than the scalar polarizability, can be either averaged out by performing measurements for all the M components or suppressed by using slightly weaker probe light intensity [23].

The differential BBR shift at the room temperature, $T = 300$ K, due to the $E1$ channel for the clock transition is estimated using the expression

$$\delta E_{\mathrm{BBR}}^{E1} = -\frac{1}{2}(831.9\ \mathrm{V/m})^2\left[\frac{T(K)}{300}\right]^4\delta\alpha_0^{E1}, \qquad (7)$$

and found to be much lower than 10^{-19} level in all four HCIs. Similarly, the BBR shift due to the other dominant $M1$ channel of the $|\eta\rangle$ state can be estimated using the formula

$$\Delta E_{\mathrm{BBR}}^{M1} = -\frac{\mu_0(K_BT)^2}{\pi^2(c\hbar)^3}\sum_\beta\Bigg[|\langle\eta||O^{M1}||\beta\rangle|^2\omega_{\eta\beta} \times \int_0^\infty d\omega\frac{\omega^3}{(\omega_{\eta\beta}^2-\omega^2)(\exp^{\hbar\omega/K_BT}-1)}\Bigg], \qquad (8)$$

where μ_0, K_B, $\hbar$, and c are the magnetic permeability, Boltzmann's constant, Planck's constant, and speed of light, respectively. The respective expressions of $\langle\eta||O^{M1}||\beta\rangle$ for the atomic and hyperfine levels are used from Eqs. (2) and (4) to estimate the $M1$ BBR shifts of the states involved with the clock transitions. Then, the BBR shift of the clock transition is determined as

$$\delta E_{\mathrm{BBR}}^{M1} = \Delta E_{\mathrm{BBR}}^{M1}(^3P_0) - \Delta E_{\mathrm{BBR}}^{M1}(^3P_2), \qquad (9)$$

which is found to be extremely small at the room temperature in all four HCIs.

The electric quadrupole shift, $\Delta E_\Theta = -\Theta\mathcal{E}_{zz}/2$ caused by the gradient of the applied electric field $\mathcal{E}_{zz}$ in the z direction on a state with quadrupole moment Θ, could be a major systematic effect in the atomic clocks. Our choice of upper state as $^3P_0$ and lower state as $^3P_2$ $(F = 1/2)$ in Cu^{13+} warrants $\Delta E_\Theta = 0$ since $\Theta = 0$ in these states. However, the ΔE_Θ values in the lower clock states in the Ni^{12+}, Pd^{12+}, and Ag^{13+} ions are substantially large due to nonzero values of Θ. It is possible to nullify such electric quadrupole shifts by performing measurements in all their M components and averaging out as has been demonstrated in [23].

In conclusion, the Ni^{12+}, Cu^{13+}, Pd^{12+}, and Ag^{13+} ions are found to be potential candidates for making optical atomic clocks with the fractional uncertainty below 10^{-19} level and the quality factor larger than 10^{15}. The atomic energy levels in these ions are simple for performing experiments and calculations. They also have magnetic dipole transitions in the optical region that can be used for laser cooling and detecting the quantum jumps on the clock transitions by the shelving method rather than using the quantum logic spectroscopy. These ions are useful for probing possible variation of the fine-structure constant by comparing their clock frequencies with astrophysical observations. Notably, they can be produced using the presently available experimental facilities.

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