## Hydrogenlike Highly Charged Ions for Tests of the Time Independence of Fundamental Constants

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Hyperfine transitions in the electronic ground state of cold, trapped hydrogenlike highly charged ions have attractive features for use as frequency standards because the majority of systematic frequency shifts are smaller by orders of magnitude compared to many microwave and optical frequency standards. Frequency measurements of these transitions hold promise for significantly improved laboratory tests of local position invariance of the electron and quark masses.

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A fascinating question in physics is whether the fundamental constants depend on space or time [1]. Either dependency would represent a violation of local position invariance and require modification of general relativity. Many astronomical studies have been performed in the past years [2], and a recent work indicates a possible time dependence of the electron-to-proton mass ratio [3]. An important complementary approach in space-time variability searches is the use of laboratory frequency standards [4]. The recently developed cold-atom frequency standards have been used for improving the upper limits for a present-day drift of several constants by more than an order of magnitude [5,6].

To improve the limits further, it is important to develop frequency standards of higher accuracy. Recently, ion optical standards with a systematic error in the  $10^{-17}$  range have been reported [7,8], lower than in the best microwave standards. While tests of local position invariance of  $\alpha$  will directly benefit from the new ion optical clocks, unfortunately these are only very weakly sensitive to constants other than  $\alpha$ . In order to address the strong and weak interactions, a sensitivity to  $m_e/m_p$  is desirable. Theoretical models suggest a significantly higher rate of change for it compared to  $\alpha$ , if the latter is not constant [9]. The currently most precise approach to test the constancy of  $m_e/m_p$  is a comparison of a microwave hyperfine transition in a cold-atom clock with an optical atomic transition. This yielded an upper limit of  $7 \times 10^{-15}/\text{yr}$ , assuming other constants not to be varying [5]. The improvement of this limit will in the future be limited by the performance of the microwave clocks. It is therefore important to develop optical standards that use transitions of different character [10]. One possibility is vibrational transitions in cold trapped molecules [11], which depend on  $m_e/m_p$  (and  $\alpha$ ) [12].

Here we propose and analyze another atomic system, hydrogenlike highly charged ions (HHCIs) (a single electron and a nucleus of charge +Ze,  $Z\gg 1$ ). The hyperfine splitting (HFS) transitions have several special features: (i) Their frequencies depend on several dimensionless fundamental and nuclear constants, including  $\alpha$ ,  $m_e/m_p$ ,

and nuclear gyromagnetic ratios. The latter two can be expressed as ratios of the masses  $m_e$ ,  $m_q$ ,  $m_s$  of the electron, the up or down quark, and the strange quark relative to the QCD energy scale  $\Lambda$  [13]. (ii) The natural Q factor of the transitions exceeds  $10^{13}$  for ions with Z less than about 70 [14]. Together with the high transition frequency, this satisfies a necessary condition for reaching very low frequency instability, required for the proposed studies. (iii) Systematic effects are very small due to the strong binding of the 1s electron to the nucleus and the spherical symmetry of the wave function. (iv) Although not laser-coolable, hydrogenlike highly charged ions can be cooled sympathetically [15].

The recently demonstrated "quantum logic" spectroscopy method [8] appears suited also for high-precision spectroscopy of HHCIs: Here a single laser-coolable atomic ion ("logic" ion of charge  $q_L$ ) and a single non-laser-coolable spectroscopy ion are trapped together in a linear radio frequency ion trap and are cooled to near rest. A transition of the sympathetically cooled ion can be read out efficiently without the need of possessing a cycling transition.

We therefore consider a generic HHCI of total charge q=(Z-1)e, mass m, and half-integer nuclear spin I trapped together with a "logic" ion, such as  ${}^9\mathrm{Be}^+$ . A small constant quantization magnetic field  $\mathbf{B}_0$  along z is assumed to be present. The clock laser's magnetic field  $B_{\mathrm{clock},z}\hat{\mathbf{z}}$  at frequency  $\nu_{\mathrm{clock}} = E_H/h$  probes the hyperfine transition  $|F=I-1/2, m_F=0\rangle \leftrightarrow |F'=I+1/2, m_{F'}=0\rangle$  between the (first-order) magnetic field-independent magnetic sublevels m=0 of the 1s state.

Berkeland *et al.* [16] demonstrated a highly accurate microwave HFS frequency standard based on trapped, laser-cooled, singly charged ions and evaluated many systematic effects. Here we extend their evaluation to the HHCI. We discuss the influence of magnetic and electric fields originating from the trap fields, the logic ion, the clock laser, the cooling laser, blackbody radiation (BBR) of the trap environment at a temperature  $T_{\rm BBR}$ , and vacuum residual pressure. The effects are summarized in Table I for a particular medium-Z and a high-Z HHCI. These were

TABLE I. Systematic frequency shifts estimates. Pairs of values refer to Ni and Pb, respectively. LS: Light shift. Assumptions: absence of excess micromotion; in (4), complete asymmetry and 10% population in the m=1 state; in (3f) and (4), large polarization misalignment  $(B_{\text{clock},x}/B_{\text{clock},z})^2 = 0.1$ .  $B_{\text{clock},z} = (0.07, 1.6)$  nT is adapted to the upper level lifetime and to the clock laser linewidth (1 Hz). The cooling laser intensity is attenuated by  $10^4$  during HHCI interrogation.

Type		$^{61}$ Ni <sup>27+</sup> ( $I = 3/2$ )	$^{207}\text{Pb}^{81+}(I=1/2)$	Parameters
	$\nu_{ m clock}$ , natural linewidth	8.9 THz, 0.2 mHz	294 THz, 3 Hz	
(1a)	BBR Stark shift	$-2 \times 10^{-18} \text{ Hz}$	$-5 \times 10^{-20} \text{ Hz}$	$T_{\rm BBR} = 10 \; {\rm K}$
(1b)	LS due to cooling laser E field	$-8 \times 10^{-19} \text{ Hz}$	$-2 \times 10^{-20} \text{ Hz}$	$E_{L_{\rm ex}} = 10^{-2} (87 \text{ V/m})$
(1c)	LS due to clock laser E field	$-5 \times 10^{-22} \text{ Hz}$	$-6 \times 10^{-21} \text{ Hz}$	$E_{\text{clock},x} = (0.02, 0.5) \text{ V/m}$
(1d)	Trap Stark shift	$-2 \times 10^{-18} \text{ Hz}$	$-2 \times 10^{-20} \text{ Hz}$	$E_{\text{trap,rms}} = (1.1, 0.65) \text{ V/m}$
(2)	Electric quadrupole shift	$1 \times 10^{-7} \text{ Hz}$	$1 \times 10^{-9} \text{ Hz}$	$(6.0, 5.7) \times 10^7 \text{ V/m}^2$
(3a)	dc 2nd order Zeeman shift	$4 \times 10^{-6} \text{ Hz}$	$9 \times 10^{-8} \text{ Hz}$	$B_{\rm dc} = 0.3 \ \mu \rm T$
(3b)	ac 2nd order Zeeman shift	$2 \times 10^{-12} \text{ Hz}$	$5 \times 10^{-14} \text{ Hz}$	$B_{\rm line} = 0.3  \rm nT$
(3c)	Trap ac 2nd order Zeeman shift	$5 \times 10^{-7} \text{ Hz}$	$1 \times 10^{-8} \text{ Hz}$	$B_{\Omega} = 0.15 \ \mu \text{T}$
(3d)	LS due to cooling laser B field	$-2 \times 10^{-14} \text{ Hz}$	$-4 \times 10^{-13} \text{ Hz}$	$B_{L,z} = 10^{-2}(0.3 \ \mu\text{T})$
(3e)	BBR Zeeman shift	$3 \times 10^{-10} \text{ Hz}$	$6 \times 10^{-12} \text{ Hz}$	$T_{\rm BBR} = 10 \text{ K}$
(3f)	LS due to clock laser B field	$4 \times 10^{-15} \text{ Hz}$	$5 \times 10^{-14} \text{ Hz}$	BBR
(4)	Line pulling	$3 \times 10^{-8} \text{ Hz}$	$6 \times 10^{-5} \text{ Hz}$	
(5)	Pressure shift	-0.04  MHz/Pa	-0.9  MHz/Pa	He gas; $T = 10 \text{ K}$
(6)	2nd order Doppler shift	$-6 \times 10^{-5} \text{ Hz}$	$-6 \times 10^{-4}$ Hz	$(T_{s,\text{clock}}, T_{s,L}) = (3, 0.01) \text{ mK}$

chosen for concreteness only; at the present stage of analysis, all half-integer nuclei with hyperfine transition frequencies above  $\sim 10$  THz appear to be suitable, in principle. HHCI with high Z could be favored because the clock laser linewidth will more likely not be a limitation to the realized Q factor.

- (1) Stark shifts.—The electric field-induced change in the 1s HFS has been calculated in Ref. [17] and measured on H and alkali atoms. The relative Stark shift scales as  $Z^{-6}$ . The shifts due to the electric fields from BBR and cooling or clock lasers can be estimated by considering the rms field strength, since the relevant frequencies are small compared to the lowest-energy electric dipole transition [18]. The shifts are negligible. The rms trap electric field strength associated with thermal motion of a single trapped ion has been evaluated by Berkeland *et al.* [19] and can be generalized to the two-ion case. In the absence of micromotion, the field is  $E_{\text{trap,rms}}^2 \simeq 2mk_BT_s\Omega^2/q^2$ , where  $T_s$  is the secular temperature, and  $\Omega$  is the trap rf angular frequency. Even allowing for significant excess micromotion, the trap Stark shift is still negligible.
- (2) Quadrupole shift.—The two hyperfine levels have small electric quadrupole moments stemming from the nuclear quadrupole moment (if present) and from admixture of the *d* state [20]. The latter contribution is smaller than typical nuclear quadrupole moments. The quadrupole moment differences are about 7–9 orders smaller than for, e.g., the clock ion Hg<sup>+</sup>, leading to negligible shifts.
- (3) Zeeman shifts.—The dc quadratic Zeeman shift of the HFS transition is given by the generalized Breit-Rabi equation [21]. The Zeeman coefficient for near-infrared HFS frequencies is of order  $\delta \nu/B^2 \sim 0.02~\text{Hz/G}^2$ , small compared to, e.g., 0.7 Hz/G² for Al<sup>+</sup> and  $-190~\text{Hz/G}^2$  for Hg<sup>+</sup>. dc magnetic fields occur from the quantization,

Earth, and equipment fields (adding to  $B_{\rm dc}$ ), while ac fields arise from line modulation (amplitude  $B_{\rm line}$ ) and the trap rf field (amplitude  $B_{\Omega}$ ). We take experimental values reported by Berkeland *et al.* [16] to obtain the shifts. Light shifts [22] are caused by the BBR and the cooling and clock lasers' magnetic fields. The dominant contribution is from BBR. For HFS transition wavelengths sufficiently short compared to the BBR peak, the quasistatic approximation together with the Breit-Rabi equation can be used.

Using the light shift theory, the effects of the clock laser and far detuned cooling laser can be calculated. The relative shifts are of order  $(\mu_B B_r / E_H)^2$ , where  $B_r$  denotes appropriate spatial components of the laser wave amplitudes  $B_{\rm clock}$  and  $B_L$ . Since these components can be minimized by proper clock laser polarization alignment and by attenuating the cooling laser during HHCI interrogation, the net shifts are tiny.

- (4) Line pulling.—A shift of the resonance may occur due to off-resonant excitation of the  $m=0 \to \pm 1$  transitions by the small field component  $B_{\mathrm{clock},x}$  and, for  $I \ge 3/2$  nuclei, additionally of the  $m \to m$ ,  $m \ne 0$  transition by  $B_{\mathrm{clock},z}$  if  $m \ne 0$  sublevels are populated. The pulling is estimated by considering the addition of the line shapes. Following Ref. [16], an upper limit can be set by assuming pulling only by the transitions with, e.g., positive detuning and 10% population of, e.g., the m=1 sublevel.
- (5) Pressure effects.—Collisions with background gas molecules or atoms lead to HCI loss by charge transfer (electron capture) to it. Little is known about the HCI capture rate from thermal or subthermal neutral particles. Cross section measurements for near-thermal energy were performed for Si<sup>4+</sup> [23] and Ar<sup>+q</sup> in charge states up to q = 6 [24], while for HCI (Xe<sup>(35-46)+</sup> and Th<sup>(73-80)+</sup>) the lowest collision energies studied were ~6q eV [25].

Ab initio calculations indicate that at thermal energies cross sections are approximately given by the Langevin value  $\sigma_L$ , which scales proportional to q [26]. This predicts the lifetime of a HCI in even the highest charge states to be tens of seconds in a cryogenic vacuum (pressure below 10<sup>-10</sup> Pa). In a cryogenic Penning trap, lifetimes  $\sim$ 50–100 s for the above HCIs have indeed been observed [15,25]. At present, there is no complete theory of HFS frequency shift for HHCIs. We expect that, for collisions with few-electron neutrals such as He and H<sub>2</sub>, a precise ab initio treatment should be feasible by extending related treatments [23,26,27]. The (negative) shift caused by the interaction of sufficiently distant neutrals interacting with an ion via the induced dipole is well known [28] and can be used as a simple model also for HHCIs. We make the plausible assumption that this interaction is limited to ranges exceeding the Langevin capture  $\sqrt{\sigma_L/2\pi}$ , since for smaller ones electron transfer takes place and the ion is lost. In this case, we find that the relative frequency shift is at the  $10^{-18}$  level or less in a cryogenic vacuum.

(6) Second-order Doppler effect.—This effect is, in principle, independent of the ion charge and scales inversely with the mass. If the radial modes of the two-ion system with a large contribution from the HCI are assumed to have a temperature  $T_s$  close to the Be<sup>+</sup> Doppler cooling limit, the calculated Doppler shift is in the  $10^{-18}$  range for heavy ions. Allowing for noncompensated excess micromotion, this shift could be 1 order larger, several parts in  $10^{-17}$ .

The transition energy between the F = I - 1/2and F = I + 1/2 hyperfine levels of the 1s state is given by  $E_H \simeq 2\alpha^4 Z^3 g_I (m_e/m_p)(2I+1)(1-\delta)(1-\epsilon)m_e c^2/\epsilon$  $3\gamma(2\gamma-1)$ , where  $\gamma=\sqrt{1-(\alpha Z)^2}$ , and  $\delta$  and  $\epsilon$  are the nuclear charge and magnetization distribution corrections, respectively [29]. Here we neglect the radiative contributions [30]. Since for most HCIs,  $\epsilon < 0.02$ , we will neglect it here.  $\delta$  attains its largest values for heavy HCIs, on the order of 0.1. Its dependence on the fundamental constants should be considered in the present context but has not yet been calculated. The dependences of the nuclear g factor and the proton mass on the masses  $m_u$ ,  $m_d$ ,  $m_s$  of the u, d, squarks can be approximately calculated [13]. The relative sensitivity of the transition energy on the various fundamental constants can be written as  $\delta E_H/E_H =$  $K_{\alpha}(Z)\delta\alpha/\alpha + K_{e}\delta\tilde{m}_{e}/\tilde{m}_{e} + K_{q}\delta\tilde{m}_{q}/\tilde{m}_{q} + K_{s}\delta\tilde{m}_{s}/\tilde{m}_{s},$ where the tilde stands for normalization to  $\Lambda$  and  $m_q =$  $(m_u + m_d)/2$ . The dimensionless coefficients K are as follows.  $K_e = 1$  and  $K_\alpha$  varies monotonically from 1 for Z = 1 to 4.3 at Z = 92. Within the Schmidt model of nuclear moments,  $(K_q, K_s) = (-0.037 - 0.486 f_N/g_I)$  $-0.011 - 0.073 f_N/g_I$ ) for nuclei with a valence proton (odd Z, even A), and  $(K_q, K_s) = (-0.037 + 0.451 f_N/g_I)$  $-0.011 + 0.005 f_N/g_I$ ) for nuclei with a valence neutron (even Z, odd A). The function  $f_N = 1/2$  for j = l + 1/2, and  $f_N = -j/(2j+2)$  for j = l - 1/2, where l and j are the orbital and total angular momentum of the valence nucleon, respectively. A more sophisticated analysis of the dependence of the nuclear moments on the quark masses has recently been given [31], which includes the effects of nuclear polarization by the valence nucleon. It was shown that for certain nuclei this reduces the magnitudes of  $K_a$ ,  $K_s$  strongly. In the framework of the present discussion, it is sufficient to point out the need to calculate these sensitivities for a large number of long-lived or stable isotopes to provide a selection guide for experiments aimed specifically at a search for a time dependence of the quark masses. While the sensitivities to the quark masses are relatively low compared to  $K_{\alpha}$  and  $K_{e}$  (also according the more accurate model), they are still useful, especially in view of the potential high accuracy of HHCI frequency standards.

For tests of constancy of fundamental constants, the dimensionless ratio  $r_{12} = E^{(1)}/E^{(2)}$  of two transition frequencies of dissimilar clocks is measured. The relative sensitivity of  $r_{12}$  to variations of any fundamental constant is determined by the differences  $K_i^{(1)} - K_i^{(2)}$  of the respective sensitivities. If we consider a comparison between HHCI frequency standards only, the ratios r contain no dependence on  $m_e/m_p$ , but the remaining three fundamental constants  $\alpha$ ,  $m_a$ ,  $m_s$  can be tested for. In order to rule out accidental (near-)cancellations of the sensitivities, at least four HCI should be intercompared, yielding three frequency ratios. The independence of the ratios can be maximized by choosing appropriate HCIs based on more accurate quark mass sensitivity calculations. Among the chosen HCIs, at least two should exhibit a large nuclear charge difference so as to obtain a strong sensitivity on  $\alpha$ .

In order to also constrain the variation of  $m_e/\Lambda$ , an additional optical standard should be used, based on an atomic electronic transition (where  $K_e=0$  and  $K_\alpha$  is atom-dependent) or on a molecular vibrational transition (where  $K_e\approx 1/2$  for fundamental vibrational transitions and  $K_\alpha=2$ ). Thus, five or more standards could test the variation of all four constants discussed here. These standards could all be atomic ion standards.

HCIs have been extensively studied [32]. Light HHCIs can be produced by electron impact ionization, accelerators, electron cyclotron resonance, or electron-beam ion sources. Precision measurements on cold, trapped light HHCIs have been performed [33]. Heavy HHCIs, up to hydrogenlike uranium, can be produced by heavy-ion accelerators and electron-beam ion traps (EBITs) [34]. The latter have the advantage of being laboratory devices. The extraction of HCIs from an EBIT, transport to an ion trap, and sympathetic cooling have already been demonstrated [15]. The HFS of HHCIs has so far been measured only for some heavy ions [35]. Studies of medium-Z HHCI included, e.g., the Lamb shift [36]. Research programs on HCIs in a cryogenic Penning trap (HITRAP project [37]) and in a cryogenic electrostatic storage ring (CSR [38]) are under development in Germany. From HITRAP, data on electron capture cross sections at low temperature should become available. The development of a HHCI frequency reference requires a suitable source that quickly and repeatedly delivers new ions, e.g., via an accumulation trap. HITRAP or the CSR could serve this purpose (with appropriate extensions) and deliver HHCIs to a linear radio frequency trap for further cooling and quantum logic spectroscopy.

In conclusion, (near-)optical frequency references based on single trapped ultracold HHCIs appear highly suitable for tests of local position invariance of fundamental constants and, in particular, of those that are not accessible using the current optical frequency standards based on singly ionized or neutral atoms. Most systematic effects are negligible at cryogenic temperature, the second-order Doppler shift and possibly the pressure shift being the dominant contributions. Assuming uncertainties to be tens of percent of the respective shifts, the frequency uncertainty could be less than several parts in  $10^{17}$ . Comparisons among appropriately selected HHCI and conventional optical standards spanning several years should then permit testing the time independence of the electron and up or down quark masses relative to the QCD energy scale at the level of  $1 \times 10^{-17}$ /yr and of the strange quark mass at the level of  $1 \times 10^{-16}$ /yr, significantly improving present limits. The practical implementation of these standards will be challenging but poses no fundamental problems and should become feasible within ongoing research programs. Another aspect of the present analysis is the perspective of determining with unprecedented accuracy the nuclear properties (albeit entangled with QED effects) of a large fraction of all stable and longlived nuclei [39].

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