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Testing the spatial variation of the fine-structure constant α indicated in [Webb et al., Phys. Rev. Lett. 107, 191101 (2011)] with terrestrial laboratory atomic measurements requires at least $\dot{\alpha}/\alpha \sim 10^{-19}$ y^{-1} sensitivity. We conduct a systematic search of atomic systems for such a test that have all features of the best optical clock transitions leading to possibility of the frequency measurements with fractional accuracy on the level of 10^{-18} or better and have a factor of 100 extra enhancement of α-variation in comparisons to experimental frequency ratio measurement accuracy. We identify the pair of actinide Cf^{15+} and Es^{16+} ions as the best system for a test of spatial $\alpha-$variation hypothesis as it satisfies both of these requirements and have sufficiently simple electronic structure to allow for high-precision predictions of all atomic properties required for rapid experimental progress.

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Theories aimed at unifying gravity with other fundamental interactions [1, 2] suggest variation of the fundamental constants, such as fine-structure constant $\alpha = e^2/\hbar c$ that characterises the strength of the electromagnetic interaction. In 2011, a very large analysis of the quasar absorption spectra which combined data taken by Keck telescope in Hawaii and the Very Large Telescope (VLT) in Chile indicated 4σ spatial gradient in the value of α [3]. Such result, implying that α was larger in the past in one direction, but smaller in the opposite direction, would have a profound significance for the "fine-tuning" problem: the values of fundamental constants that can support life happen to fall within a narrow range [4, 5]. 2015 study of systematic distortions in the wavelengths scales of high-resolution spectrographs [6] showed that instrumental error may weaken the spatial variation result [3], but can not explain all of the observed variation of α. Therefore, it became paramount to set up a laboratory experiment capable of potential observation of the spatial α-variation at the level indicated by [3].

Measurement of an oscillating evolution of α may be used to search for a low-mass primordial dark matter field [7, 8],transient effects may indicate passing of the cosmic topological defects [9, 10].

Frequencies of atomic transitions depend on α, therefore long-term monitoring of frequency ratio with ultra-high precision atomic clocks enables tests of α-variation at the present time. We also note a special case of non-clock test of α-variation in Dy [11], which uses extremely high sensitivity of a specific transition between two almost degenerate excited states while accuracy of the frequency measurements is significantly lower than in atomic clocks. Current laboratory limit for time variation of the fine structure constant stands on the level of 2×10^{-17} fractional change per year [11–14]. However, test the spatial alpha-dipole hypothesis presented in [3] requires at least two orders of magnitude improvement in accuracy of laboratory measurements. The rate at which α is changing in time due to Earth movement in the framework of alpha-dipole is [5]

$$\frac{1}{\alpha}\frac{\partial \alpha}{\partial t} = (1.35\,\cos\psi + 0.014\,\cos\omega t) \times 10^{-18}\ \mathrm{yr}^{-1}, \quad (1)$$

where $\cos\psi \sim 0.07$ is related to the direction of the Sun movement relative to the alpha-dipole, and second term in (1) is due to Earth rotation around the Sun.

The worlds best optical lattice atomic clocks approach fractional accuracy of 10^{-18} [15–17], with the smallest uncertainty of 2×10^{-18} achieved for Sr [15]. However, this does not immediately translates into high sensitivity to α-variation, because Sr and Yb clock atomic transitions are not sufficiently sensitive to the variation of the fine structure constant [18].

The dependence of atomic frequencies on α can be parameterized by the sensitivity coefficient q [19, 20]

$$\omega(x) = \omega_0 + qx, \quad (2)$$

which can be rather accurately determined from theoretical computations. Here,

$$x = \left[\left(\frac{\alpha}{\alpha_0}\right)^2 - 1\right]$$

and α_0 is a current value of α.The parameter q links variation of frequency ω to the variation of α

$$\frac{\delta\omega}{\omega_0} = \frac{2q}{\omega_0}\frac{\delta\alpha}{\alpha_0} \equiv K\frac{\delta\alpha}{\alpha_0}, \quad (3)$$

where $K = 2q/\omega_0$ is an enhancement factor. In atomic clock laboratory α-variation tests, the ratio of two clock frequencies is monitored, and the sensitivity of such test is then described by the difference in their respective K values, i.e. $\Delta K = |K_2 - K_1|$:

$$\frac{\partial}{\partial t} \ln \frac{\omega_2}{\omega_1} = (K_2 - K_1) \frac{1}{\alpha} \frac{\partial \alpha}{\partial t} \qquad (4)$$

The K factor for Sr clock transitions is 0.06 [21]. In fact, the K factors for most of the clocks currently in development, Mg, Al^+, Ca^+, Sr^+, Sr, Yb, Yb^+ quadrupole transition, and Hg are very small, $0.008 - 1.0$ [21]. The K factors for Hg^+ clock and Yb^+ octupole clock transitions are -2.9 and -6, making them the best candidates for one part of the clock comparison pair against the clock from the previous group that will essentially serve as a reference with little sensitivity to α-variation. As a result, reaching $\dot{\alpha}/\alpha \sim 10^{-20}$ y^{-1} precision with any of these clock combinations would require better than 10^{-19} precision of the clock ratio, even with one of the clocks based on octupole Yb^+ transitions. Comparison of different clocks will become even more challenging beyond 10^{-18} accuracy due to sensitivity to the environment, including temperature and gravitational potential [22]. For example, a clock on the surface of the Earth that is higher by just 1 cm than another identical clock runs faster by $\delta\omega/\omega_0 \sim 10^{-18}$ [22].

Therefore, it is highly desirable to find a combination of two transitions which can be used to design of a high-precision clocks and has at least $\Delta K = 100$, which is a subject of this work. The transitions which combine high sensitivity to α-variation with potential to design very accurate optical clock can be found in highly charged ions (HCI) [23–26] or in a unique case of ^{229}Th nucleus [27]. The use of the HCI for search of the time variation of the fine structure constant α ($\alpha = e^2/\hbar c$) was considered in Refs. [23–25, 28–32]. Sympathetic cooling of HCI ions with Be^+ has just been demonstrated in [33], paving the way to the future clock development.

We have studied a large variety of highly-charged ions systems to identify the systems with $\Delta K > 100$ that approximately satisfy the criteria for good clock transitions formulated in Ref. [26]:

- The transition is in optical region (230 nm $< \lambda <$ 2000 nm or 5000 $\mathrm{cm}^{-1} < \hbar\omega <$ 43000 cm^{-1}).
- The lifetime of the clock state is between 100 and $\sim 10^4$ seconds.
- There are other relatively strong optical transitions (equivalent lifetime on the order of $\tau \lesssim 1$ ms).
- Clock transition is not sensitive to perturbations caused by the black-body radiation (BBR), gradients of external electric fields, etc.

The largest sensitivity of atomic optical transitions to the variation of the fine structure constant has been found so far in Cf^{16+} ions [24]. However, the states of Cf^{16+} sensitive to variation of α lack important features of clock states listed above. First four excited states of Cf^{16+} ion are metastable, however, the lifetimes of all these clock states are outside of the desirable value range, which will make accurate measurements problematic. In the present work, we find that the pair of Cf^{15+} and Es^{16+} ions satisfy the clock requirements with the enhanced sensitivity to α-variation of $\Delta K = 110$. Both of these ions has first excited metastable state which has all features of the upper clock state, such as convenient values of the frequencies and transition rates, low sensitivity to external perturbations, etc. The transition between ground and these metastable states corresponds to the $f - p$ single-electron transition. This makes the transitions sensitive to the variation of the fine-structure constant. An additional advantage comes from the fact that there is a $5f - 6p$ level crossing between Cf $^{15+}$ and Es^{16+} ions, i.e. the ground state of Cf^{15+} becomes upper clock state i n Es^{16+} and vice versa. This means that if α changes in time, the drift of clock frequencies in Cf^{15+} and Es $^{16+}$ have different signs, e.g. if α increases, the clock frequency of Cf^{15+} increases as well while clock frequency of Es^{16+} decreases. This leads to extra enhancement of the sensitivity of the ratio of clock frequencies to the variation of the fine structure constant.

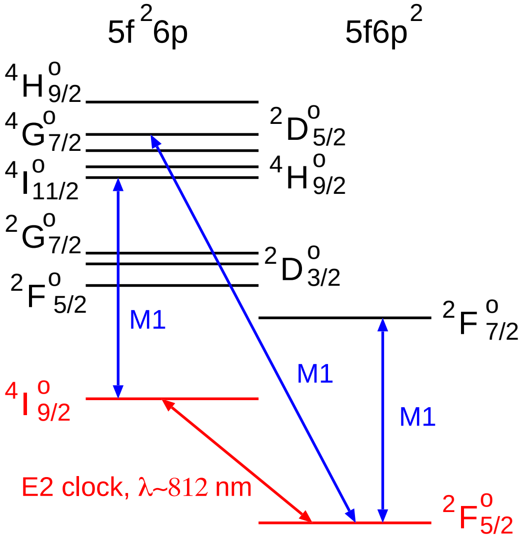

FIG. 1: Low-lying energy levels of Cf^{15+} (roughly in scale). Leading configurations are shown on the top. Clock transition is the electric quadrupole (E2) transition between the ground and first excited state. Strongest magnetic dipole (M1) transitions are also shown.

Another advantage of using Cf^{15+} and Es^{16+} ions is due to the fact that they represent good compromise between simple electron structure and the abundance of relatively strong optical transitions which can be used for cooling, detection, etc. Simple electron structure allows for reliable theoretical predictions of the ion properties, which will tremendously simplify the spectra identification and experimental search for the clock transitions. Most of the systems considered before have either simple electron structure [23–25, 29–31] but not many strong op-

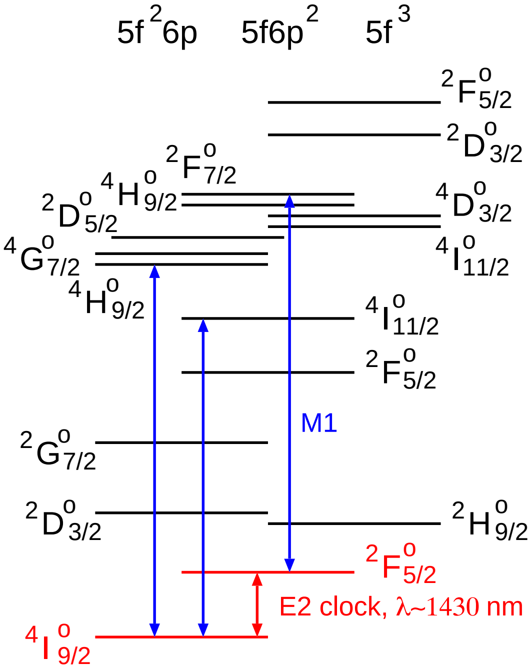

FIG. 2: Low-lying energy levels of Es^{16+}, see caption for Fig. 1.

tical transitions, or complicated electron structure (many electrons in open shells) [26, 28, 32] that leads to experimental difficulties.

Neither californium nor einsteinium have stable isotopes. However, they have very long-lived isotopes, such as ^{249}Cf with half life time of 351 years and ^{252}Es with half life time of 1.3 years. There are many facilities around the globe (Berkeley, Dubna, Darmstadt, RIKEN, etc.) which produce and study unstable isotopes (see, e.g. [34, 35]).

Below, we described our predictions for the properties of these ions relevant to the search for α-variation. We use the CI+all-order method [36, 37], which combines the single-double linearised coupled-cluster [38] and the configuration interaction approaches. The B-spline technique [39] is used to construct a set of single-electron basis states in the V^{N-3} approximation [40], which means that the initial self-consistent Hartree-Fock procedure is done for the Hg-like closed-shell core with three valence electrons removed. Core-valence and core-core correlations are included with the use of the single-double coupled cluster method while valence-valence correlations between the three electrons are included with the use of the configuration interaction method. Breit and quantum electrodynamic (QED) corrections are also included as described in Refs. [41, 42].

Judging by previous experience with the CI+all-order method [36, 37, 43, 44] we expect the error in calculated energies to be within few per cent of the ionization potential value. The analysis of the accuracy of present calculations is not simple due to absence of experimental data.

TABLE I: Excitation energies (in cm^{-1}), g-factors and lifetimes (in s) of the lowest states of Cf^{15+} and Es^{16+} ion. The uncertainties for the energies do nor exceed 500 cm^{-1} for the states of $5f6p^2$ configuration and can be almost two times larger for the states of $5f^26p$ and $5f^3$ configurations. The uncertainty for lifetimes is between 10 and 50%. The numbers in brackets represent powers of 10.

Ion	Conf.	Term	Energy	g-factors	Lifetime
Cf^{15+}	$5f6p^2$	$^2\mathrm{F}^o_{5/2}$	0	0.843	
	$5f^26p$	$^4\mathrm{I}^o_{9/2}$	12314	0.813	6.9[+3]
	$5f6p^2$	$^2\mathrm{F}^o_{7/2}$	21947	1.083	1.2[-2]
	$5f^26p$	$^2\mathrm{F}^o_{5/2}$	26665	0.715	0.26
	$5f^26p$	$^2\mathrm{D}^o_{3/2}$	27750	0.765	1.9
	$5f^26p$	$^2\mathrm{G}^o_{7/2}$	28875	0.868	1.6[-2]
	$5f^26p$	$^4\mathrm{I}^o_{11/2}$	36564	0.996	3.1[-3]
	$5f^26p$	$^4\mathrm{H}^o_{9/2}$	37392	1.029	1.5[-2]
Es^{16+}	$5f^26p$	$^4\mathrm{I}^o_{9/2}$	0	0.808	
	$5f^26p$	$^2\mathrm{F}^o_{5/2}$	6994	0.788	1.6[+4]
	$5f^3$	$^2\mathrm{H}^o_{9/2}$	10591	0.804	3.4
	$5f^26p$	$^2\mathrm{D}^o_{3/2}$	11056	0.806	666
	$5f^26p$	$^2\mathrm{G}^o_{7/2}$	15441	0.842	0.70
	$5f^26p$	$^2\mathrm{F}^o_{5/2}$	22616	0.778	0.17
	$5f^26p$	$^4\mathrm{I}^o_{11/2}$	24301	0.990	3.1[-3]
	$5f^26p$	$^4\mathrm{H}^o_{9/2}$	28351	1.026	1.2[-2]

We have estimated the uncertainties by analysing small contributions such as QED corrections, higher-order correlation corrections, the effect of basis truncation, etc. The analysis show that the uncertainty for the energies of the most states do not exceed 500 cm^{-1}. For some states of the $5f^3$ configurations it may go up to 900 cm^{-1}. The uncertainty for amplitudes of strong transitions is within 10% and can be significantly larger for weak transitions. The relative uncertainty for transition probabilities and lifetimes is significantly larger than for the energies due to their strong dependence on transition frequency. E.g., the uncertainty for the M1 transition is at least three times larger than that for the energy (see, Eq. 6), and uncertainty for the E2 transition is at least five times larger than for the energy (see, Eq. 5). The sensitivity coefficients q are more stable in the calculations than the small frequency ω. Corresponding uncertainty is within few per cent. The accuracy of the analysis can be further improved when the frequencies are measured.

The results for Cf^{15+} and Es^{16+} ions are presented in Table I. Energy level diagrams for these ions are shown in Figs. 1 and 2, respectively. Atomic properties of the clock transitions in Cf^{15+}, Es^{16+}, and Es^{17+} ions are listed in Table II. We also included Es^{17+} ion properties in Table II since it also satisfies the clock requirements, but has lower sensitivity to α-variation.

Clock transitions in Es^{17+}, Cf^{15+} and Es^{16+} ions are electric quadrupole transitions (E2) between ground and first excited state (see Figs. 1 and 2). Probability of spontaneous emission from the clock to the ground state

TABLE II: Clock transitions in Cf^{15+}, Es^{16+} and Es^{17+}. α_s is scalar static polarisability (a.u.), λ is the clock transition wavelength, β is relative frequency shift due to BBR, τ is the lifetime of the clock state, ω is the frequency of the clock transition, q is the the coefficient of sensitivity to α-variation, K is enhancement factor. Estimated uncertainty is smaller than 10% for the energies and enhancement factor K, few percent for sensitivity coefficients q, 10 to 20% for polarizabilities α_s and BBR frequency shift parameter β, and up to 50% for lifetimes. The numbers in brackets represent powers of 10.

Ion	Clock transition						τ	λ	$\hbar\omega$	β	q	K
	Ground state		α_s	Clock state		α_s	s	nm	cm^{-1}			
$^{249}Cf^{15+}$	$5f6p^2$	$^2F^o_{5/2}$	0.317	$5f^26p$	$^4I^o_{9/2}$	0.183	6900	812	12314	-2.9[-18]	380000	57
$^{253}Es^{16+}$	$5f^26p$	$^4I^o_{9/2}$	0.161	$5f^26p$	$^2F^o_{5/2}$	0.199	16000	1430	6994	1.6[-18]	-184000	-53
$^{252}Es^{17+}$	$5f^2$	$^3H_4$	0.032	$5f6p$	$^3F_2$	0.042	11000	1343	7445	4.0[-19]	-46600	-13

TABLE III: Strong magnetic dipole (M1) transitions involving ground state (GS) and clock state (CS) of Cf^{15+}, Es^{16+} and Es^{17+} ions. A is the amplitude in Bohr magnetons μ ($\mu = e\hbar/2mc$), τ is the equivalent lifetime, which is the inverse of the rate of spontaneous emission from upper state to clock or ground state. See Table II and text for the discussion of uncertainties.

Ion	Lower State	Upper State	Wavelength nm	Frequency (cm^{-1})	A (μ)	τ (ms)
Cf^{15+}	GS	$^2F^o_{7/2}$	456	21947	1.55	11
	CS	$^4I^o_{11/2}$	412	24250	3.17	3
Es^{16+}	GS	$^4I^o_{11/2}$	412	24301	3.03	3
	CS	$^2F^o_{7/2}$	359	27837	1.82	4
Es^{17+}	GS	$^3H_5$	476	21014	2.98	5

is given by (atomic units)

$$T^{\rm E2}_{c\rightarrow g} = \frac{1}{15}(\alpha\omega)^5\frac{\langle c||E2||g\rangle^2}{2J_c+1}, \quad (5)$$

where α is the fine structure constant, ω is the frequency of the transition, and J_c is the total angular momentum of the clock state. Corresponding lifetimes are presented in Table II. Note that they are either within or very close to the desirable range.

All considered here actinide ions have relatively strong magnetic dipole transitions to the ground and clock states. Note that since we are considering ions in the vicinity of the $6p-5f$ level crossing only states of the same parity lie within optical range. Therefore, the strongest optical transitions are always M1 transitions. The rate of spontaneous emission from upper state a to lower state b is given by (atomic units)

$$T^{\rm M1}_{a\rightarrow b} = \frac{4}{3}(\alpha\omega)^3\frac{\langle a||M1||b\rangle^2}{2J_a+1}. \quad (6)$$

The strongest M1 amplitudes are presented in Table III.

Frequency shift due to black body radiation can be presented in a form

$$\delta\omega/\omega \approx \beta(T/T_0)^4, \quad (7)$$

where T is temperature, T_0 is room temperature (T_0= 300 K), and parameter β is given by

$$\beta = -\frac{\Delta\alpha_s}{2\omega}\left(831.9\ {\rm V/m}\right)^2. \quad (8)$$

TABLE IV: Long-living isotopes of Cf and Es with convenient values of nuclear spin (I).

Isotope	Lifetime	I
^{249}Cf	351 y	9/2
^{252}Es	1.29 y	5
^{253}Es	20 d	7/2
^{255}Es	40 d	7/2

Here $\Delta\alpha_s$ is the difference in static scalar polarizabilities of clock and ground states, ω is the frequency of the clock transition.

Calculated polarizabilities and BBR shift parameter β for considered here clock transitions are presented in Table II. Note that the fractional BBR shift is of the the order of 10^{-18} even at 300 K. It can be further reduced by lowing the temperature. The clock states considered in this work have large value of the total angular momentum J (see Table II). As a result, they have non-zero quadrupole moment that couples to gradient of external electric fields. However, corresponding frequency shift can be avoided by choosing appropriate components of the hyperfine structure of the clock states. The quadrupole energy shift is proportional to $3M^2-F(F+1)$, where F is total atomic angular momentum including nuclear spin I ($\mathbf{F}=\mathbf{J}+\mathbf{I}$), M is projection of F on z-axis. The shift is zero for states with $F=3$, $M=2$ or $F=0$, $M=0$. It can also be numerically suppressed for appropriate hyperfine structure states [45]. Table IV lists isotopes of Cf and Es which have long lifetime and suitable values of nuclear spin. Using the ^{249}Cf isotope for the Cf^{15+} clock, the ^{252}Es isotope for the Es^{17+} clock and ^{253}Es or ^{255}Es isotopes for the Es^{16+} clock makes it always possible to chose states with $F=3$, $M=2$ for both clock states. For upper state of $^{249}Cf^{15+}$ clock the choice of $F=0$, $M=0$ is also possible.

The results for q and K parameters of clock transitions are presented in Table II. Note that q values are very large, with the different sign of q for Cf and Es ions. The $6p$ - $5f$ transition in the Cf^{15+} ion becomes the $5f$

- $6p$ transition in the Es^{17+} and Es^{16+} ions, thus reversing the sign of the K. While the leading configurations for both clock states of the Es^{16+} ion are the same the sensitivity coefficient q is still large. This results from the strong admixture of the $5f6p^2$ configuration, which is different for the lower and upper states. If the ratio of the $^{253}Es^{16+}$ clock frequency and the $^{249}Cf^{15+}$ clock frequency is measured, the sensitivity is given by

$$\frac{\partial}{\partial t}\ln\frac{\omega_2}{\omega_1} = 110\frac{1}{\alpha}\frac{\partial\alpha}{\partial t}. \quad (9)$$

according to Eq. 4. Comparing this with (1) shows that sensitivity to detect alpha-dipole can be achieved if frequencies are measured with relative accuracy of $\sim 10^{-18}$.

In summary, we find that the pair of Cf^{15+} and Es^{16+} ions satisfy the requirements for the design of a high-precision clock with the enhanced sensitivity to α-variation of $\Delta K = 110$. Therefore, if α changes in time or with spatial position, the ratio of the Cf^{15+} and Es^{16+} clock frequencies changes more than one hundred times faster. This drastically reduces the stringent requirement to the accuracy of the frequency measurement needed to achieve sensitivity for the test of the spatial α-variation indicated by the astrophysical observations [3]. We have surveyed a large number of potential systems and large K was either due to a small transition frequency ω or occurred in systems that make design of accurate clock difficult due to other reason, such as too short or too long clock state lifetimes. To the best of our knowledge, this is the only pair of ions with such a large δK that also satisfy the clock requirements introduced in [26].

We note that Cf^{15+} is a particular good clock candidate with optical (812 nm) clock transition, very long-lived isotope that conveniently allows for $M = 0$ $F = 0$ selection of the clock state, and very large $K = 57$. Therefore, an experiment measuring ratio of Cf^{15+} clock frequency with any other current clock where 10^{-18} accuracy can be achieved will also provide excellent test of α-variation, which will lose only factor of two in sensitivity to Cf/Es pair but would be easier to implement.

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