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Atomic optical clocks and search for variation of the fine-structure constant

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Theories unifying gravity and other interactions suggest the possibility of spatial and temporal variations of physical ''constants.'' Accuracy achieved for atomic optical frequency standards (optical clocks) approaches the level when possible time evolution of the fine-structure constant α can be studied by comparisons of rates between clocks based on different atomic transitions in different atoms. The sensitivity to variation of α is due to relativistic corrections that are different in different atoms ($\sim Z^2\alpha^2$). We have calculated the values of the relativistic energy shifts in In II, Tl II, Ba II, and Ra II, which all can be used as atomic optical clocks. The results are to be used to translate any change in the clock's rate into variation of α.

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Possible variations of the fundamental physical constants are suggested by unified theories, such as string theory and *M* theory (see, e.g., [1–3]). A number of works have been done in the past few years in an attempt to find experimental evidence of any space-time variation of the fine-structure constant α. The search goes mostly in two ways. One is based on analysis of the absorption spectra of distant quasars. Comparing the spectra of atoms or ions in distant gas clouds, which intersect the sight lines towards the quasars with the laboratory spectra, allows one to put bounds on the space-time variation of α. Another way uses precise atomic clocks in laboratory measurements. Different atomic transitions depend differently on the fine-structure constant. Comparing the rates of different atomic clocks over long periods of time allows one to put bounds on the local change of α with time. Astrophysical measurements have a big advantage of having a many-orders-of-magnitude enhancement factor gained by looking into the distant past. At the present time the strongest bound on the possible space-time variation of α has been obtained from the analysis of the quasar absorption spectra. There is even evidence that the value of α might be smaller in early epochs [4]. However, the accuracy achieved for atomic clocks now approaches the level where measurements with similar accuracy become possible. These measurements are also important because they produce results that are independent of the cosmological model and any possible space variation of the fundamental constants.

The strongest laboratory limit on the time variation of α was obtained by comparing H-maser vs Hg II microwave atomic clocks over 140 days [5]. The Fermi-Segre formula for the hyperfine splitting and the Casimir relativistic correction factor were used to translate frequency drift into a variation of α. This yielded an upper limit $\dot{\alpha}/\alpha \leqslant 3.7\times 10^{-14}$/yr.

Another possibility is to use optical atomic frequency standards. These standards are based on strongly forbidden $E1$ transitions or $E2$ transitions between the ground state of an atom (ion) and its close metastable excited state. Proposed optical frequency standards include Ca I [6], Sr II [7], Yb II [8], Hg II [9], Mg I [10], In II [11], Xe I [12], Ar I [13], etc. In contrast with the microwave frequency standards, there is no simple analytical formula for the dependence of optical atomic frequencies on α. This dependence can be revealed via accurate relativistic calculations only. In our earlier paper [14] we presented such calculations for Ca I, Sr II, Yb II, and Hg II. We stress that relativistic corrections cannot be reduced to spin-orbit interaction. For example, the *s*-electron level has the largest relativistic correction and no spin-orbit interaction [14].

Recently an experiment to measure a possible time variation of α was proposed in Ref. [15] by linking H and In II optical frequency standards. In the present work we calculate the relativistic energy shift of the clock transition of In II and its heavier analog Tl II. For the search for variation of α Tl II may be preferable since it has larger relativistic effects (the relative magnitude of the relativistic corrections increases as $Z^2\alpha^2$ with the nuclear charge Z). We also include in the calculations some other metastable states of both ions. Ba II ion had been considered as a candidate for an optical frequency standard in Ref. [17]. Therefore, we perform the calculations for Ba II and its heavier analog Ra II as well.

It is convenient to represent the results in the form

$$\omega = \omega_0 + q_1 x + q_2 y, \tag{1}$$

where $x = (\alpha/\alpha_l)^2 - 1$, $y = (\alpha/\alpha_l)^4 - 1$, and ω_0 is an experimental frequency of a particular transition. To find the value of the coefficients q_1 and q_2 we have repeated the calculations for $\alpha = \alpha_0$, $\alpha = \sqrt{7/8}\alpha_0$, and $\alpha = \sqrt{3/4}\alpha_0$ and fit the results by formula (1). We started the calculations from the relativistic Hartree-Fock method. The V^{N-1} approximation was used to generate a complete set of core and valence basis states. Correlations between the core and valence electrons have been included by means of many-body perturbation theory. In the cases of In II and Tl II, which both have two valence electrons above the closed-shell core, the correlations between the valence electrons have been included by means of the configuration interaction method. A more detailed discussion of the methods of calculation can be found in our earlier work [14].

The obtained values of the coefficients q_1 and q_2 as well as experimental frequencies of some ''clock'' transitions in In II, Tl II, Ba II, and Ra II are presented in Table I. Note that

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TABLE I. Relativistic energy shift of the ${}^1S_0$-${}^3P_0$ clock transition of In II and some ground-to-metastable-states transitions of In II, Tl II, Ba II, and Ra II (cm^{-1}) [see formula (1) for the definition of q_1 and q_2].

Z	Ion	Ground state		Upper states		ω_0 [a]	q_1	q_2
49	In II	$5s^2$	${}^1S_0$	$5s5p$	${}^3P_0$	42275	2502	956
				$5s5p$	${}^3P_1$	43349	3741	791
				$5s5p$	${}^3P_2$	45827	6219	791
81	Tl II	$6s^2$	${}^1S_0$	$6s6p$	${}^3P_0$	49451	1661	9042
				$6s6p$	${}^3P_1$	53393	5877	8668
				$6s6p$	${}^3P_2$	61725	14309	8668
56	Ba II	$6s$	${}^2S_{1/2}$	$5d$	${}^2D_{3/2}$	4843.850	5402	221
				$5d$	${}^2D_{5/2}$	5674.824	6872	−448
88	Ra II	$7s$	${}^2S_{1/2}$	$6d$	${}^2D_{3/2}$	12084.38	15507	1639
				$6d$	${}^2D_{5/2}$	13743.11	19669	−864

[a] Moore, Ref. [21].

$$\dot{\omega}|_{\alpha=\alpha_0}=(2q_1+4q_2)\frac{\dot{\alpha}}{\alpha}. \tag{2}$$

The most recent and strongest limits on the time variation of α are following:

$\dot{\alpha}/\alpha<3.7\times10^{-14}/\mathrm{yr}$	Prestage *et al.* [5]
$\dot{\alpha}/\alpha<10^{-15}/\mathrm{yr}$	Damour and Dyson [18]
$\dot{\alpha}/\alpha<10^{-15}/\mathrm{yr}$	Webb *et al.* [4]
$\dot{\alpha}/\alpha<1.9\times10^{-14}/\mathrm{yr}$	Ivanchik *et al.* [19]

Only first of these results is a local present-day limit on the time variation of α. It was obtained by comparing Hg II and H microwave atomic clocks as was mentioned before. The second result was obtained from analysis of the Oklo natural nuclear reactor. The Oklo event took place in Gabon (Africa) around 1.8×10^9 years ago. The two other results came from the analysis of the astrophysical data and correspond to even larger time intervals. It is interesting to see what accuracy is needed to improve the present-day limit on variation of α. Substituting $\dot{\alpha}/\alpha=10^{-14}$ and q_1 and q_2 from Table I into formula (2) we can get for the In II clock transition

$$\dot{\omega}=2.6\ \mathrm{Hz/yr}\quad(\dot{\alpha}/\alpha=10^{-14}\ \mathrm{yr}^{-1}),\quad \mathrm{In\ II}. \tag{3}$$

Note, that the natural linewidth of the clock line of In II is 1.1 Hz [15]. The frequency of the ^{115}In II clock transition is currently known to an accuracy $\sim10^{-11}$:

$$\omega_0=1\,267\,402\,452\,914(41)\ \mathrm{kHz}$$

(see Refs. [15] and [16]). However, further improvement in accuracy up to the level of $\sim10^{-15}$ is probably possible [20]. In fact, it is enough to measure the variation of the ratio or the difference between two frequencies. Note, that the relativistic energy shift of the ${}^1S_0$-${}^3P_0$ transition in Tl II is about five times larger:

$$\dot{\omega}=12\ \mathrm{Hz/yr}\quad(\dot{\alpha}/\alpha=10^{-14}\ \mathrm{yr}^{-1}),\quad \mathrm{Tl\ II}. \tag{4}$$

Relativistic effects are also large for upper metastable states of In II and Tl II and for the s-d transitions in Ba II and Ra II (see Table I). In principle, all these states can be used for atomic clocks.

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