

**Relativistic effects in Sr, Dy, Yb II, and Yb III and search for variation of the fine-structure constant**

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 (Received 14 May 2003; published 21 August 2003)

A possibility for fundamental constants to vary in time is suggested by theories unifying gravity with other interactions. In this article we examine proposals to use optical transitions of Sr, Dy, Yb II, and Yb III for the search of the time variation of the fine-structure constant  $\alpha$ . Frequencies of atomic transitions are calculated using the relativistic Hartree-Fock method and configuration interaction technique. The effect of variation of  $\alpha$  on the frequencies is studied by varying  $\alpha$  in computer codes. Accuracy of measurements needed to improve the current best limit on the time variation of  $\alpha$  is discussed.

DOI: 10.1103/PhysRevA.68.022506

PACS number(s): 31.30.Jv, 06.20.Jr, 31.15.Ar

**I. INTRODUCTION**

Theories unifying gravity with other interactions suggest that fundamental constants could vary in space-time (see, e.g., [1]). Recent evidence of variation of the fine-structure constant  $\alpha$  in quasar absorption spectra [2] elevated interest to the search of variation of  $\alpha$  in laboratory experiments. Comparing frequencies of different atomic transitions over a long period of time is a good way to do such a search due to the extremely high accuracy of measurements achieved for certain types of transitions. The best limit on local present-time variation of the fine-structure constant published so far was obtained by comparing the  $\text{Hg}^+$  microwave atomic clock versus hydrogen maser [3]. Recently this limit was further improved by more than an order of magnitude in comparing cesium and rubidium atomic clocks [4]. There are also many proposals for the search of a variation of  $\alpha$  in atomic optical transitions, some of which were analyzed in our previous works (see [5] and references therein). In the present paper, we analyze three new proposals involving strontium or calcium, dual beam [6], dysprosium atom [5,7], and ytterbium positive ions  $\text{Yb}^+$  [8] and  $\text{Yb}^{2+}$  [9]. We perform relativistic many-body calculations to link the variation of  $\alpha$  with the variation of the frequencies of atomic transitions. Then we use this connection to find out what accuracy of measurements is needed to improve the current best limit on time variation of the fine-structure constant.

In the proposal suggested by Bergeson, the strontium-calcium dual beam is to be used to compare the frequencies of the  $^1S_0$ - $^3P_1$  clock transitions in these atoms over a long period of time. Ca and Sr have similar electron structure. However, due to higher nuclear charge, relativistic effects are larger for strontium. If  $\alpha$  is changing, the corresponding change in frequency of the clock transition for Sr would go considerably faster than for Ca. Precise measurements might be able to indicate this or, at least, put a strong constraint on a possible variation of  $\alpha$ . Calculations of the relativistic effects for Ca were done in our previous work [5]. In the present paper, we do similar calculations for Sr.

Experiments with a ytterbium positive ion have the advantage of greater relativistic effects due to larger nuclear charge and the convenience of working with two different transitions of the same element. There are two transitions in  $\text{Yb}^+$  involving metastable states for which comparison of

frequencies is considered. One is a quadrupole transition  $4f^{14}6s^2S_{1/2}$ - $4f^{14}5d^2D_{5/2}$  and the other is an octupole transition  $4f^{14}6s^2S_{1/2}$ - $4f^{13}6s^2F_{7/2}$ . The quadrupole transition is basically an  $s$ - $d$  transition while the octupole one is an  $f$ - $s$  transition. According to the simple analytical formula presented in Ref. [5], relativistic energy shifts for  $s$  electrons and electrons with high total momentum  $j$  (such as  $d$  and  $f$  electrons) are large but have opposite sign. This means that we should expect that two metastable states of  $\text{Yb}^+$  move in opposite directions if  $\alpha$  is changing. This brings extra enhancement to the sensitivity of the measurements for  $\text{Yb}^+$  to the variation of  $\alpha$ . Our accurate calculations presented below support these considerations.

The proposal for dysprosium is quite different from what was considered so far. Instead of comparing two very stable atomic clock frequencies, the authors of this proposal [5] suggest measuring a very small frequency of the transition between two almost degenerate states of opposite parity in dysprosium. The states are  $4f^{10}5d6s^3[10]_{10}$   $E=19797.96\text{ cm}^{-1}$  and  $4f^95d^26s^9K_{10}$   $E=19797.96\text{ cm}^{-1}$ . These states were used before for the search of parity nonconservation in Dy [11]. Small energy splitting and different electron structure of these two states lead to very strong enhancement of the sensitivity of the frequency of transition between the states to variation of  $\alpha$ . The enhancement (about eight orders of magnitude) seems to be strong enough to overcome the disadvantage of dealing with states which are not very narrow.

In the present paper, we calculate the values of relativistic energy shifts for Sr,  $\text{Yb}^+$ , and Dy and discuss what accuracy of measurements is needed to improve the current best constraint on local time variation of the fine-structure constant.

**II. CALCULATIONS OF ENERGIES**

We use relativistic Hartree-Fock (RHF) and configuration interaction (CI) methods to do the calculations.

The RHF Hamiltonian is used to generate a set of single-electron orbitals. We use a form of single-electron wave function which explicitly depends on the fine-structure constant  $\alpha$ ,

$$\psi(r)_{njlm} = \frac{1}{r} \begin{pmatrix} f_n(r)\Omega(\mathbf{n})_{jlm} \\ i\alpha g_n(r)\tilde{\Omega}(\mathbf{n})_{jlm} \end{pmatrix}. \quad (1)$$

Then the RHF equation for  $\psi(r)_n$  has the following form (in atomic units):

$$f'_n(r) + \frac{\kappa_n}{r} f_n(r) - [2 + \alpha^2(\epsilon_n - \hat{V}_{HF})] g_n(r) = 0,$$

$$g'_n(r) - \frac{\kappa_n}{r} f_n(r) + (\epsilon_n - \hat{V}_{HF}) f_n(r) = 0, \quad (2)$$

where  $\kappa = (-1)^{l+j+1/2}(j+1/2)$ ,  $n$  is the principal quantum number, and  $\hat{V}_{HF}$  is the Hartree-Fock potential. The value of relativistic effects is studied by varying the value of  $\alpha$  in Eq. (2). In particular, the nonrelativistic limit corresponds to  $\alpha = 0$ .

In the CI calculations, we use an approach similar to what was first developed in Ref. [12]. Electrons occupying open shells are considered as valence electrons and all others are core electrons. Correlations between valence electrons are treated within the CI method while correlations between valence and core electrons are included by means of the many-body perturbation theory. The effective CI Hamiltonian for valence electrons is presented by

$$\hat{H}^{CI} = \sum_{i=1}^N \hat{h}_i + \sum_{i<j} \frac{e^2}{r_{ij}}. \quad (3)$$

Here  $N$  is the number of valence electrons and  $\hat{h}_i$  is an effective single-electron Hamiltonian,

$$\hat{h}_i = c \boldsymbol{\alpha} \cdot \mathbf{p} + (\beta - 1) m c^2 - \frac{Z e^2}{r_i} + \hat{V}_{\text{core}} + \hat{\Sigma}_1. \quad (4)$$

Here  $V_{\text{core}}$  is the Hartree-Fock potential created by core electrons. It differs from  $V_{HF}$  in Eq. (2) by contribution of valence electrons.  $\hat{\Sigma}_1$  is the so-called ‘‘correlation potential’’ operator. It describes correlations between a particular valence electron and core electrons (see Ref. [12] for details). Note that in contrast with Ref. [12], we do not include in the present work the  $\hat{\Sigma}_2$  operator, which is a two-electron operator describing a different type of correlations between valence and core electrons. Terms with  $\hat{\Sigma}_2$  can be considered as screening of the Coulomb interaction between valence electrons by core electrons. These terms are less important than those with  $\hat{\Sigma}_1$  but much more time consuming in calculations. We either neglect them or simulate their effect by introducing screening factors.

We are now going to discuss the specifics of the calculations for each atom/ion. Apart from the states of interest, we also calculate energies of the other states of the same configurations to ensure that the accuracy is systematically good. We also calculate magnetic  $g$  factors to ensure correct identification of states. This is particularly important for dysprosium.

### A. Strontium

Strontium in its ground state is a closed-shell atom. It has two  $5s$  electrons on its outermost shell and we need to con-

sider energy intervals between the  $^1S_0$  ground state and states of the  $5s5p$  configuration where the  $^3P_1$  metastable state is of most interest. The RHF calculations for Sr were done in the  $V^N$  approximation for a closed-shell atom in its ground state. For the CI calculations we considered Sr as an atom with two valence electrons and followed similar calculations for Ba [13]. Basis states for the CI+MBPT method were calculated using the B-spline technique [14] with 40 B-splines in a cavity of radius  $R = 40a_B$ . The same basis functions were used to calculate  $\hat{\Sigma}_1$  for the CI calculations. Thirteen lowest states above the core in each of the  $s_{1/2}$ ,  $p_{1/2}$ ,  $p_{3/2}$ ,  $d_{3/2}$ , and  $d_{5/2}$  waves were used to construct a two-electron wave function for both  $5s^2$  and  $5s5p$  configurations. A large number of basis functions is needed mostly for an adequate description of the  $5s5p$  configuration. This is because the  $V^N$  approximation does not provide us with a good  $5p$  single-electron state. Also, the  $5s$  single-electron state in the  $5s5p$  configuration is different from the  $5s$  state in the  $5s^2$  configuration for which Hartree-Fock calculations were done. However, with 13 states in each wave the saturation of the basis was clearly achieved and adding more states to the basis did not change the energy. Two-electron basis states for the CI calculations were obtained by distributing valence electrons over 65 basis states ( $13 \times 5$ ) in all possible ways with a restriction of fixed parity and total momentum.

The results are presented in Table I. As one can see, accuracy for the state of interest  $^3P_1$  is better than 1% while accuracy for other states is also good.

### B. Ytterbium

The ground state of a ytterbium positive ion is  $4f^{14}6s^2S_{1/2}$  and we need to consider transitions into the  $4f^{14}5d^2D_{5/2}$  and  $4f^{13}6s^2F_{7/2}$  states. Therefore, it is convenient to do the RHF calculations in the  $V^{N-1}$  approximation for the  $\text{Yb}^{2+}$  ion with the  $4f^{14}$  closed-shell configuration. The  $6s, 5d$  and other basis states for the CI method are calculated then in the field of the frozen closed-shell core of  $\text{Yb}^{2+}$ . Then, in the CI calculations, we need to consider all  $4f$  electrons as valence ones since one of the transitions of interest involves excitation from the  $4f$  subshell. So, the total number of valence electrons in the present CI calculations is 15. This is very different from our previous calculations for  $\text{Yb}^+$  [5] in which the  $4f^{13}6s^2F_{7/2}$  state was not considered and we were able to treat the ytterbium ion as a system with one external electron above closed shells.

Our final set of single-electron states for the CI calculations consisted of  $4f_{5/2}$ ,  $4f_{7/2}$ ,  $6s_{1/2}$ ,  $5d_{3/2}$ ,  $5d_{5/2}$ , and a few more  $s$  and  $f$  states above  $4f$  and  $6s$ . Note that in contrast with Sr, we do not need many basis functions here because all our single-electron wave functions correspond to the  $\text{Yb}^+$ . This makes the initial approximation very good and leads to fast convergence of the CI calculations with respect to the basis set used.

We also do not include  $\hat{\Sigma}_1$  in calculations for  $\text{Yb}^+$ . In a case of many valence electrons (15 for  $\text{Yb}^+$ ), correlations are dominated by correlations between them which are taken into account accurately via the CI technique. Correlations between valence electrons and core electrons mostly mani-

TABLE I. Energies and  $g$  factors.

Atom/ion	State		Energy (1/cm)		$g$		Nonrelativistic
			Theory	Experiment <sup>a</sup>	Theory	Experiment <sup>a</sup>	
Sr	$5s^2$	$^1S_0$	0	0			
	$5s5p$	$^3P_0$	14171	14318			
	$5s5p$	$^3P_1$	14384	14504			
	$5s5p$	$^3P_2$	14832	14899			
	$5s5p$	$^1P_1$	22829	21698			
Dy	$4f^{10}6s^2$	$^5I_8$	0	0	1.243	1.242	1.25
	$4f^{10}6s^2$	$^5I_7$	4123	4134	1.175	1.173	1.179
	$4f^{10}6s^2$	$^5I_6$	7147	7051	1.072	1.072	1.071
	$4f^{10}6s^2$	$^5I_5$	9428	9212	0.907	0.911	0.9
	$4f^{10}6s^2$	$^5I_4$	11199	10925	0.614	0.618	0.6
	$4f^{10}5d6s$	$^3[8]_9$	18605	17515	1.319	1.316	
	$4f^{10}5d6s$	$^3[9]_{10}$	18615	18463	1.291	1.282	
	$4f^{10}5d6s$	$^3[10]_{11}$	19811	19349	1.268	1.27	
	$4f^{10}5d6s$	$^3[10]_{10}$	20133	19798	1.208	1.21	
	$4f^95d^26s$	$^9K_{12}$	23345	22541	1.327	1.333	1.333
	$4f^95d^26s$	$^9K_{11}$	20513	20448	1.352	1.354	1.303
	$4f^95d^26s$	$^9K_{10}$	19623	19798	1.372	1.367	1.264
	$4f^95d^26s$	$^9I_9$	19434	19558	1.390	1.39	1.377
	$4f^95d^26s$	$^9G_8$	18379	18473	1.461	1.46	1.5
	$4f^95d^26s$	$^9G_7$	18662	18529	1.492	1.467	1.5
	$4f^95d^26s$	$^7F_6$	19714	19304	1.527	1.54	1.5
	$4f^95d^26s$	$^7G_5$	21697	20892	1.510	1.32	1.37
	$4f^95d^26s$	$^9G_4$	23748	22697	1.492	1.487	1.5
Yb <sup>+</sup>	$4f^{14}6s$	$^2S_{1/2}$	0	0	2.000	1.998	2
	$4f^{14}5d$	$^2D_{3/2}$	22888	22961	0.800	0.800	0.8
	$4f^{14}5d$	$^2D_{5/2}$	23549	24333	1.200	1.202	1.2
	$4f^{13}6s^2$	$^2F_{5/2}$	31820	31568	0.857	0.862	0.857
	$4f^{13}6s^2$	$^2F_{7/2}$	21819	21419	1.143	1.145	1.143
	$4f^{14}6p$	$^2P_{1/2}$	26000	27062	0.667	0.667	0.667
	$4f^{14}6p$	$^2P_{3/2}$	29005	30392	1.333	1.333	1.333

<sup>a</sup>References [16,17].

fest themselves via screening of the Coulomb interaction between valence electrons. We take this effect into account semiempirically by introducing screening factors  $f_k$ . Namely, we multiply every Coulomb integral of the multipolarity  $k$  by a numerical factor  $f_k$  which is chosen to fit the energies. It turns out that a good fit for Yb<sup>+</sup> is achieved with  $f_2=0.8$  and  $f_k=1$  for all other  $k$ .

Many-electron basis states for the CI calculations were obtained by allowing all possible single and double excitations from the base configuration with the restriction of fixed parity and total momentum.

Results for energies of Yb<sup>+</sup> are presented in Table I. The theoretical accuracy for energies as compared to the experiment is 2–3 % for the states of interest and is not worse than 5% for other states.

### C. Dysprosium

The dysprosium atom is the most difficult for calculations because of its complicated electron structure. Ground-state configuration of Dy is  $4f^{10}6s^2$ , which means that there is no

realistic RHF approximation which corresponds to a closed-shell system. We do the RHF calculations for Dy in the  $V^N$  approximation with an open-shell version of the RHF method. Contribution of the  $4f$  electrons into the RHF potential is calculated as for a closed shell and then multiplied by a numerical factor to take into account its fractional occupancy. This factor is 10/14 when interaction of the  $4f$  electrons with other core electrons is considered and 9/13 when interaction of a  $4f$  electron with other  $4f$  electrons is considered. When convergence is achieved, we have the  $4f$  and  $6s$  basis states for the CI calculations. To calculate other states of valence electrons we remove one  $6s$  electron, freeze all RHF orbitals, including  $4f$  and  $6s$ , and calculate the  $6p_{1/2}$ ,  $6p_{3/2}$ ,  $5d_{3/2}$ ,  $5d_{5/2}$  and a few more  $d$  states above  $5d$  in the field of frozen RHF core.

In the CI calculations, states below  $4f$  are considered as core states and all others as valence states. The total number of valence electrons is therefore 12. As for the case of Yb<sup>+</sup>, we neglect  $\hat{\Sigma}_1$  and use screening factors as fitting parameters to improve agreement with experiment. It turns out that the

TABLE II. Relativistic energy shift  $q(\text{cm}^{-1})$ .

Atom/ion	State		This work	Ref. [5]
Sr	$5s5p$	$^3P_1$	667	
	$5s5p$	$^1P_1$	1058	
Dy	$4f^{10}5d6s$	$^3[10]_{10}$	6008	
	$4f^95d^26s$	$^9K_{10}$	-23708	
Yb II	$4f^{14}5d$	$^2D_{3/2}$	10118	12582
	$4f^{14}5d$	$^2D_{5/2}$	10397	11438
	$4f^{13}6s^2$	$^2F_{7/2}$	-56737	
Yb III	$4f^{13}5d$	$^3P_0$	-27800	

best fit for the  $4f^{10}6s5d$  configuration is achieved with  $f_1 = 0.7$  and  $f_k = 1$  for all other  $k$ . No fitting was used for other configurations.

To calculate states of the  $4f^{10}6s^2$ ,  $4f^{10}6s6p$ , and  $4f^{10}6s5d$  configurations, we use the  $4f_{5/2}$ ,  $4f_{7/2}$ ,  $6s_{1/2}$ ,  $6p_{1/2}$ ,  $6p_{3/2}$ ,  $5d_{3/2}$ , and  $5d_{5/2}$  single-electron basis functions and all possible configurations which can be obtained from these basis functions by exciting one or two electrons from the base configuration. The same approach does not work for the  $4f^95d^26s$  configuration because of the huge number of many-electron basis states generated this way, and as a consequence, the CI matrix is of so large a size that it could not be handled by our computers. On the other hand, test calculations with pairs of configurations showed that mixing of our state of interest with other configurations is small and can be neglected. We do need, however, to include mixing with the  $4f^95d6d6s$ ,  $4f^95d7d6s$ , and  $4f^96d^26s$  configurations. This is because our basis  $5d$  state corresponds rather to the  $4f^{10}5d6s$  configuration and extra  $d$  states are needed to correct it.

The results are presented in Table I. Note that they are considerably better than in our previous calculations [15]. This is because of the better basis and more complete CI treatment.

### III. FREQUENCY SHIFT: RESULTS AND DISCUSSIONS

In the vicinity of the physical value of the fine-structure constant ( $\alpha = \alpha_0$ ), the frequency ( $\omega$ ) of an atomic transition can be presented in the form

$$\omega = \omega_0 + qx, \quad (5)$$

where  $x = (\alpha^2/\alpha_0^2) - 1$ ,  $\omega_0$  is the experimental value of the frequency, and  $q$  is a coefficient which determines the frequency dependence on the variation of  $\alpha$ . To find the values of  $q$  for different atomic transitions, we repeat all calculations for  $\alpha = \sqrt{\frac{9}{8}}\alpha_0$ ,  $\alpha = \alpha_0$ , and  $\alpha = \sqrt{\frac{7}{8}}\alpha_0$ . Then

$$q = 4(\omega_+ - \omega_-), \quad (6)$$

where  $\omega_+$  is the value of  $\omega$  for  $\alpha = \sqrt{\frac{9}{8}}\alpha_0$  and  $\omega_-$  is the value of  $\omega$  for  $\alpha = \sqrt{\frac{7}{8}}\alpha_0$ . Calculations for  $\alpha = \alpha_0$  are done to compare the theory with experiment and to check whether frequencies are linear functions of  $\alpha^2$ . The results for coef-

ficients  $q$  are presented in Table II. Note that we have included in the table the results of our old calculations for  $\text{Yb}^+$ . These calculations were done in a very different way, assuming that  $\text{Yb}^+$  is an atom with one external electron above closed shells. Comparison of the results obtained by different methods gives an estimate of the accuracy of calculations.

A search for the time variation of the fine-structure constant can be conducted by comparing two frequencies of atomic transitions over a long period of time. The measured value can be presented as [5,3]

$$\Delta(t) = \left( \frac{\dot{\omega}_1}{\omega_1} - \frac{\dot{\omega}_2}{\omega_2} \right). \quad (7)$$

Using Eq. (5) one can reduce Eq. (7) to

$$\Delta(t) = \left( \frac{2q_1}{\omega_1} - \frac{2q_2}{\omega_2} \right) \left( \frac{\dot{\alpha}}{\alpha_0} \right). \quad (8)$$

The current best laboratory limit on the time variation of  $\alpha$  is  $\dot{\alpha}/\alpha < 10^{-15} \text{ yr}^{-1}$  [4].

In the first experiment considered in this paper, a dual calcium-strontium beam is to be used to compare the frequencies of the  $^1S_0 - ^3P_1$  transitions in both atoms. Substituting  $\omega_1 = 15\,210 \text{ cm}^{-1}$ ,  $q_1 = 230 \text{ cm}^{-1}$  for Ca [5];  $\omega_2 = 14\,504 \text{ cm}^{-1}$ ,  $q_2 = 667 \text{ cm}^{-1}$  for Sr (Tables I and II); and  $\dot{\alpha}/\alpha = 10^{-15} \text{ yr}^{-1}$  we get

$$\Delta(t)(\text{Sr}-\text{Ca}) = 6.2 \times 10^{-17} \text{ yr}^{-1}. \quad (9)$$

Note that the width of the  $^3P_1$  state in Sr may be a problem in this case.

In the case of  $\text{Yb}^+$ , frequencies of the  $^2S_{1/2} - ^2D_{5/2}$  and  $^2S_{1/2} - ^2F_{7/2}$  are to be compared. Substituting the numbers, we get

$$\Delta(t)(\text{Yb}^+) = 6.1 \times 10^{-15} \text{ yr}^{-1}. \quad (10)$$

Note the two orders of magnitude improvement in the magnitude in comparison with the Sr-Ca dual beam experiment.

We have also calculated the  $q$  coefficient for the  $4f^{14}1S_0 - 4f^{13}5d\ ^3P_0$  ( $\omega = 45\,276 \text{ cm}^{-1}$ ) transition from the Yb III ground state. This was motivated by the proposed measurements [9] of the  $\alpha$  variation using a comparison of  $^1S_0 - ^3P_0$  transition frequencies in  $\text{In}^+$ ,  $\text{Tl}^+$ , and the odd isotope of  $\text{Yb}^{2+}$ . The different signs and magnitudes of relativistic corrections in  $\text{In}^+$  ( $q = 4414 \text{ cm}^{-1}$ ),  $\text{Tl}^+$  ( $q = 19\,745 \text{ cm}^{-1}$ ), and  $\text{Yb}^{2+}$  ( $q = -27\,800 \text{ cm}^{-1}$ ) provide an excellent control of systematic errors since systematic errors are not correlated with signs and magnitudes of the frequency shifts  $qx$ , where  $x = (\alpha^2/\alpha_0^2) - 1$ . The same idea (combination of anchors, positive shifters, and negative shifters) has been used to control systematic errors in Ref. [2].

In our view, a very interesting possibility is that for dysprosium. Instead of comparing frequencies of different tran-

sitions, one should measure the energy difference between two very close states of opposite parity. The corresponding  $q$  coefficient is  $q = 6008 + 23\,708 = 29\,716 \text{ cm}^{-1}$  (see Table II). The frequency of this transition ranges from a few MHz to a few GHz depending on isotopes and hfs components used. If we take, e.g.,  $\omega = 3.1 \text{ MHz}$  [10], we get

$$\Delta(t)(\text{Dy}) = 5.7 \times 10^8 \left( \frac{\dot{\alpha}}{\alpha_0} \right). \quad (11)$$

This is an eight orders of magnitude enhancement in the relative value of the effect compared to atomic clock transitions. Substituting  $\dot{\alpha}/\alpha = 10^{-15} \text{ yr}^{-1}$ , we get

$$\Delta(t)(\text{Dy}) = 5.7 \times 10^{-7} \text{ yr}^{-1}. \quad (12)$$

This means that to improve the current best limit on local time variation of  $\alpha$ , the frequency of this transition in Dy should be measured to an accuracy of about  $10^{-7}$  over about a year time interval. This seems to be feasible [7].

#### ACKNOWLEDGMENTS

We are grateful to S. Bergeson, P. Blythe, D. Budker, S. Lea, F. Pereira, and J.R. Torgerson for useful discussions. Part of the work was done on the computers of the Australian Center for Advanced Computing and Communications. This work is supported by the Australian Research Council.

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