

## Multipolar theory of blackbody radiation shift of atomic energy levels and its implications for optical lattice clocks

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Blackbody radiation (BBR) shifts of the  $^3P_0-^1S_0$  clock transition in the divalent atoms Mg, Ca, Sr, and Yb are evaluated. The dominant electric-dipole contributions are computed using accurate relativistic many-body techniques of atomic structure. At room temperatures, the resulting uncertainties in the  $E1$  BBR shifts are large and substantially affect the projected  $10^{-18}$  fractional accuracy of the optical-lattice-based clocks. A peculiarity of these clocks is that the characteristic BBR wavelength is comparable to the  $^3P$  fine-structure intervals. To evaluate relevant  $M1$  and  $E2$  contributions, a theory of multipolar BBR shifts is developed. The resulting corrections, although presently masked by the uncertainties in the  $E1$  contribution, are required at the  $10^{-18}$  accuracy goal.

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Atomic clocks based on the ultranarrow  $^3P_0-^1S_0$  transition in divalent atoms may offer a new level of time-keeping accuracy. In this scheme the atoms are confined in an optical lattice. The lattice laser wavelength is selected in such a way that the dominant perturbations of the clock frequency, the induced ac Stark shifts, for both clock states exactly cancel. Although other effects still perturb the clock frequency, estimates [1] indicate that the projected fractional uncertainty of such clocks may be as low as  $10^{-18}$ . By comparison,  $10^{-15}$  is the fractional uncertainty of the current Cs standard realizing the SI definition of the unit of time. This advantage of the optical-lattice clocks has motivated a number of recent proposals: the original scheme of Katori [2] with fermionic Sr isotopes has been extended to Mg [3], Ca [4], and Yb [5] atoms. In addition, various schemes for probing the highly forbidden  $nsnp\ ^3P_0-ns^2\ ^1S_0$  clock transition in bosonic isotopes have been proposed: three-photon transition, electromagnetically-induced transparency, and transition assisted by a magnetic field [6–8].

Considering the advantages of optical lattice clocks, here we investigate an important systematic effect of the blackbody radiation (BBR) on the frequency of the  $^3P_0-^1S_0$  clock transition. Indeed, the SI definition of the second explicitly involves atomic clock operating at the absolute zero of temperature [9]. In a laboratory environment with an ambient temperature  $T$ , one needs to introduce the  $T$ -dependent BBR correction to the observed frequency. Even in Cs the value of the BBR shift is a subject of recent controversy [10]. Here, using techniques of many-body relativistic atomic structure, we compute the BBR shift for Mg, Ca, Sr, and Yb and evaluate uncertainties of the calculations. As summarized in Table I, the resulting fractional uncertainties in the clock frequencies at  $T=300$  K are *large*, ranging from  $1 \times 10^{-17}$  for Mg to  $3 \times 10^{-16}$  for Yb.

The main conclusions of this paper are (i) the present uncertainty in our computed BBR shift is an obstacle on the way toward the projected  $10^{-18}$  accuracy goal; (ii) due to  $T^4$  scaling of the BBR shift, it may be beneficial to operate at cryogenic temperatures; (iii) if operating at room temperatures, high-precision (0.02% accurate for Sr) measurements

of the BBR shifts or related quantities are required; (iv) the Mg-based clock is the least susceptible to BBR; compared to Sr, the Mg BBR shift is an order of magnitude smaller (see Table I). Additionally, we develop a relativistic theory of the BBR shift caused by multipolar components of the radiation field.

The paper is organized as follows. First, we derive relativistic formulas for the BBR shift which incorporate various multipolar contributions of the radiation field. Second, we apply these general expressions to determination of the BBR shifts. Unless specified otherwise, atomic units and the Gaussian system of electromagnetic units are used throughout. Temperature is expressed in units of  $E_H/k_B$ , where  $E_H$  is the Hartree energy and  $k_B$  is the Boltzmann constant.

The BBR shift is caused by perturbation of the atomic energy levels by the oscillating thermal radiation. Both atomic levels involved in the clock transition are perturbed and the overall BBR correction is the difference of the BBR shifts for the two levels. We find that the determining shift for the upper  $^3P_0$  level requires certain care. This level is a part of the  $^3P_J$  fine-structure manifold,  $J=0,1,2$ . The separation between the levels in the manifold is comparable to the characteristic wave number of the BBR radiation,  $208.51\text{ cm}^{-1}$ , at  $T=300$  K, and contributions of the BBR-induced magnetic-dipole and electric-quadrupole transitions

TABLE I. Blackbody radiation shift for clock transitions between the lowest-energy  $^3P_0$  and  $^1S_0$  states in divalent atoms.  $\delta\nu_{\text{BBR}}$  is the BBR shift at  $T=300$  K with our estimated uncertainties.  $\nu_0$  is the clock transition frequency, and  $\delta\nu_{\text{BBR}}/\nu_0$  is the fractional contribution of the BBR shift. The last column lists fractional errors in the absolute transition frequencies induced by the uncertainties in the BBR shift.

Atom	$\delta\nu_{\text{BBR}}$ (Hz)	$\nu_0$ (Hz)	$\delta\nu_{\text{BBR}}/\nu_0$	Uncertainty
Mg	-0.258(7)	$6.55 \times 10^{14}$	$-3.9 \times 10^{-16}$	$1 \times 10^{-17}$
Ca	-1.171(17)	$4.54 \times 10^{14}$	$-2.6 \times 10^{-15}$	$4 \times 10^{-17}$
Sr	-2.354(32)	$4.29 \times 10^{14}$	$-5.5 \times 10^{-15}$	$7 \times 10^{-17}$
Yb	-1.34(13)	$5.18 \times 10^{14}$	$-2.6 \times 10^{-15}$	$3 \times 10^{-16}$

to the levels of the manifold may be enhanced. Taking these induced transitions into account requires going beyond the conventional electric-dipole approximation, Ref. [11].

Considering the potential importance of the multipolar contributions, in this section we derive the relevant formulas for BBR-induced energy shifts. Although we show that the  $M1$  and  $E2$  contributions can be neglected at the present level of uncertainty for the dominant  $E1$  shift, incorporating  $M1$  multipoles will be required if the lattice clocks reach their projected  $10^{-18}$  accuracy level.

The BBR spectral density is given by the Planck formula  $u_\omega(T) = \alpha^3 \pi^{-2} \omega^3 / [\exp(\omega/T) - 1]$ . It is a weak perturbation and the time evolution of the reference state  $|g\rangle$  for off-resonance excitations can be computed assuming that the excited-state amplitudes adiabatically follow that of the reference state. With a generalization to narrow resonant contributions [11], the BBR shift is given by

$$\delta E_g = \frac{1}{4\alpha^2} \sum_{\epsilon} \int d\hat{\mathbf{k}} \text{P} \int_0^\infty u_\omega(T) \frac{d\omega}{\omega^2} \sum_p \left( \frac{[h^{(+)}]_{gp}[h^{(-)}]_{pg}}{\omega_{gp} + \omega} + \frac{[h^{(-)}]_{gp}[h^{(+)}]_{pg}}{\omega_{gp} - \omega} \right), \quad (1)$$

with averaging over photon propagation directions  $\hat{\mathbf{k}}$  and polarizations  $\epsilon$ . The second-order summation is over intermediate atomic states  $|p\rangle$  and involves the Coulomb-gauge couplings  $h^{(\pm)} = (\boldsymbol{\alpha} \cdot \boldsymbol{\epsilon}) \exp[\mp i(\mathbf{k} \cdot \mathbf{r})]$ ,  $\boldsymbol{\alpha}$  encapsulating the conventional Dirac matrices. P denotes the Cauchy principal value; as elucidated in Ref. [11] it is required for a proper treatment of resonant contributions.

We further use a multipolar expansion of  $e^{i(\mathbf{k} \cdot \mathbf{r})}$  in vector spherical harmonics and express the resulting couplings in terms of traditional multipole moments  $Q_{JM}^{(\lambda)}$ :

$$(\boldsymbol{\alpha} \boldsymbol{\epsilon}) e^{i(\mathbf{k} \cdot \mathbf{r})} = - \sum_{JM\lambda} i^{J+1+\lambda} [\mathbf{Y}_{JM}^{(\lambda)}(\hat{\mathbf{k}}) \cdot \boldsymbol{\epsilon}] \times \sqrt{\frac{4\pi(2J+1)(J+1)}{J}} \frac{k^J}{(2J+1)!!} Q_{JM}^{(\lambda)}.$$

Here  $\lambda=0$  is for magnetic (MJ) and  $\lambda=1$  is for electric (EJ) multipolar amplitudes. Relativistic expressions with retardation for matrix elements of  $Q_{JM}^{(\lambda)}$  can be found in Ref. [12]. Neglecting retardation effects,  $Q_{JM}^{(1)}$  become the frequency-independent EJ moments. In the case of magnetic-dipole transitions in the nonrelativistic limit  $Q_{1M}^{(0)} = -\frac{\alpha}{2} (\mathbf{L} + 2\mathbf{S})_M$ . Notice that the retardation brings a correction on the order of  $(\alpha\omega)^2$  to these expressions.

Averaging over polarizations and propagation directions in Eq. (1), we find that the BBR shift is a sum over multipolar contributions:  $\delta E_g = \sum_{J\lambda} \delta E_g^{(J\lambda)}$ ,

$$\delta E_g^{(J\lambda)} = -\pi \frac{J+1}{J[(2J-1)!!]^2} \alpha^{2(J-1)} \times \text{P} \int_0^\infty d\omega \omega^{2(J-1)} u_\omega(T) \alpha_g^{(J\lambda)}(\omega). \quad (2)$$

Here  $\alpha_g^{(J\lambda)}(\omega)$  are the generalized dynamic multipolar scalar polarizabilities

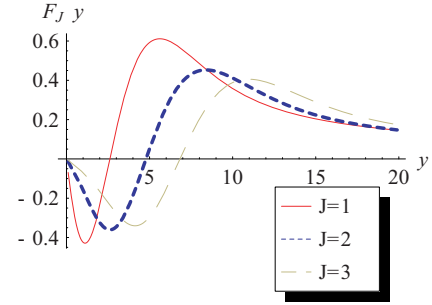


FIG. 1. (Color online) Multipolar functions  $F_J(y)$ , Eq. (5), for  $J=1, 2, 3$ .

$$\alpha_g^{(J\lambda)}(\omega) = \frac{2}{2J+1} \sum_{p,M} |\langle p | Q_{JM}^{(\lambda)} | g \rangle|^2 \left( \frac{\omega_{pg}}{\omega_{pg}^2 - \omega^2} \right). \quad (3)$$

A cursory examination of these formulas reveals that compared to the  $2^J$  multipole, the contribution of the  $2^{J+1}$  multipole is suppressed by a factor of  $\alpha^2$ . Also for the same  $J$  the magnetic contribution is  $\alpha^2$  weaker than that of the EJ photons. To illuminate the  $T$  dependence of contributions of individual intermediate states we recast the BBR shifts into the form ( $J_g$  is the total angular momentum of the reference state and  $\langle g | | Q_{JM}^{(\lambda)} | | p \rangle$  is the reduced matrix element)

$$\delta E_g^{(J\lambda)} = -\frac{(\alpha T)^{2J+1}}{2J_g+1} \sum_p |\langle g | | Q_{JM}^{(\lambda)} | | p \rangle|^2 F_J \left( \frac{\omega_{pg}}{T} \right), \quad (4)$$

with universal functions ( $x = \omega/T$ )

$$F_J(y) = \frac{1}{\pi J(2J+1)!!(2J-1)!!} \times \text{P} \int_0^\infty \left( \frac{1}{y+x} + \frac{1}{y-x} \right) \frac{x^{2J+1}}{e^x - 1} dx. \quad (5)$$

The functions  $F_J(y)$  are multipolar generalizations of function  $F(y)$  introduced by Farley and Wing [11] in the  $E1$  case. We plot our computed  $F_J$  functions for  $J=1, 2, 3$  in Fig. 1.  $F_J(y)$  are odd functions with respect to  $y$ . From examining Fig. 1, it is clear that  $F_J$  rapidly changes around  $y \sim 1$  and slowly falls off for  $y \gg 1$ . Depending on the value of the excitation energy,  $\omega_{pg} = yT$ , a particular intermediate state may introduce either a negative or a positive BBR shift. Notice that  $F_J$  are broad distributions and they have comparable values for  $|y| \lesssim 20$ ; this will have implications for interpreting our results.

At large values of the argument  $|y| \gg 1$ ,  $F_J(y) \propto 1/y$ . The limit  $y \gg 1$  corresponds to the case when the transition energy is much larger than  $T$ . If all virtual transitions satisfy this requirement, then the leading contribution to the multipolar BBR shift can be expressed in terms of static polarizabilities

$$\delta E_g^{(J\lambda)} = -\frac{\zeta(2J+2)(2J+2)!}{2\pi J[(2J-1)!!]^2} \alpha^{2J+1} T^{2J+2} \alpha_g^{(J\lambda)}(0), \quad (6)$$

where  $\zeta$  is the Riemann zeta function. As the scaling factor  $\alpha^{2J+1} T^{2J+2}$  is expressed in atomic units, we observe that as

the multipolarity  $J$  increases by 1, in addition to the usual  $\alpha^2$  suppression, there is a temperature suppression factor of  $(k_B T/E_H)^2$ . For  $T=300$  K this suppression is sizable, as  $(k_B T/E_H)^2 \approx 9.0 \times 10^{-7}$ .

Below we apply the developed formalism to computing the BBR shift for the  $^1S_0$ - $^3P_0$  clock transition in divalent atoms. We will assume that the atoms are at the ambient temperature of  $T=300$  K. Both clock levels experience the BBR shift and the total shift is the difference between the individual shifts,  $\delta\nu_{\text{BBR}} = \delta\nu_{\text{BBR}}(^3P_0) - \delta\nu_{\text{BBR}}(^1S_0)$ .

Consider first the BBR shift of the ground  $^1S_0$  state. Here the transition energies of various multipolar transitions to the upper levels are much larger than  $T$ , i.e., we are in the  $y \gg 1$  limit of Fig. 1. Here, compared to the dominant  $E1$ -induced shift, the contribution of  $M1$  transitions is suppressed by  $\alpha^2 \sim 10^{-4}$  and  $E2$  by  $\alpha^2(k_B T/E_H)^2 \sim 10^{-10}$ . Higher-order multipoles are suppressed even more. As to the retardation effects in  $E1$  matrix elements, we expect that they will be suppressed by a factor of  $\alpha^2(k_B T/E_H)^2 \sim 10^{-10}$ . Nevertheless, since the fractional contribution of the BBR shift to the clock frequency is at the  $5 \times 10^{-15}$  level (see Table I), one would need to introduce the  $M1$  corrections at the projected accuracy of  $10^{-18}$ .

For the  $^3P_0$  levels, the characteristic thermal photon frequency is comparable to the fine-structure intervals for the  $^3P_J$  manifold. The  $^3P_0$  level is connected by  $M1$  transition to the  $^3P_1$  level and by  $E2$  transition to the  $^3P_2$  level. For these transitions the values of the relevant functions  $F_J \sim 1$  (see Fig. 1), and we estimate  $\delta E_g^{(M1)} \sim \alpha^2(\alpha T)^3$ ,  $\delta E_g^{(E2)} \sim (\alpha T)^5$ , while  $\delta E_g^{(E1)} \sim \alpha^3(T)^4/\omega^3 D_{1-3P_0}$ . Our numerical estimate, based on the transitions inside the fine-structure manifold, lead to the following values of the BBR shifts for Sr:  $\delta E_g^{(M1)} = 2.4 \times 10^{-5}$  Hz and  $\delta E_g^{(E2)} = 2.5 \times 10^{-8}$  Hz. Since the  $E1$  BBR shift for Sr is  $\sim 2$  Hz, the  $M1$  and  $E2$  contributions can be neglected at the present 1% level of accuracy of our calculations.

We find that, although the thermal photon energy is close to the fine-structure intervals, the induced multipole BBR shifts are not amplified. The main reason is that the BBR energy distribution is broad: the functions  $F_J$  have comparable values for a wide range of excitation energies,  $|\omega| \lesssim 20 T$  (see Fig. 1). For example, for the Sr  $^3P_0$ - $^3D_0$   $E1$  transition  $F_1 \approx 0.16$ , while for the  $^3P_0$ - $^3P_1$   $M1$  transition  $F_1 \approx -0.41$  and for the  $^3P_0$ - $^3P_2$   $E2$  transition  $F_2 \approx -0.36$ . For such a broad distribution, the multipolar BBR shift is determined by the prefactor in Eq.(4), resulting in a suppression of multipoles beyond  $E1$ .

Based on the above discussion, we may exclusively focus on the electric-dipole ( $J=1, \lambda=1$ ) contribution to the BBR shift. From our general expressions we obtain an approximate formula

$$\delta E_g^{(E1)} \approx -\frac{2}{15}(\alpha\pi)^3 T^4 \alpha_g^{(E1)}(0)[1 + \eta],$$

TABLE II. Static electric dipole polarizabilities in a.u. for the ground  $^1S_0$  and the lowest-energy  $^3P_0$  excited states of Mg, Ca, Sr, and Yb atoms. Theoretical uncertainties are indicated in parentheses.

	Mg	Ca	Sr	Yb
$\alpha_{^1S_0}^1$	71.3(7)	157.1(1.3)	197.2(2)	111.3(5)
$\alpha_{^3P_0}^3$	101.2(3)	290.3(1.5)	458.3(3.6)	266(15)

$$\eta = \frac{(80/63)\pi^2}{\alpha_g^{(E1)}(0)T} \sum_p \frac{|\langle p || Q_1^{(1)} || g \rangle|^2}{(2J_g + 1)y_p^3} \left( 1 + \frac{21\pi^2}{5y_p^2} + \frac{336\pi^4}{11y_p^4} \right). \quad (7)$$

Here  $y_p = \omega_{pg}/T$  and  $\alpha_g^{(E1)}(0)$  is the traditional static dipole polarizability. To arrive at the above equation, we used the asymptotic expansion  $F_1(y) \approx \frac{4\pi^3}{45y} + \frac{32\pi^5}{189y^3} + \frac{32\pi^7}{45y^5} + \frac{512\pi^9}{99y^7}$ , which has an accuracy better than 0.1% for  $|y| > 10$ .  $\eta$  represents a “dynamic” fractional correction to the total shift. The leading contribution is determined by polarizability and below we compute  $\alpha_g^{(E1)}(0)$  using the methods of atomic structure.

Evaluation of the static dipole polarizabilities follows the relativistic many-body procedure of Refs. [13–15]. The employed formalism is a combination of the configuration-interaction method in the valence space with many-body perturbation theory for core-polarization effects. In this method, one determines wave functions by solving the effective many-body Schrödinger (Dirac) equation  $\{H_{\text{FC}} + \Sigma(E)\}|\Psi_n\rangle = E_n|\Psi_n\rangle$ . Here  $H_{\text{FC}}$  is the frozen-core Dirac-Hartree-Fock Hamiltonian and self-energy operator  $\Sigma$  is a core-polarization correction. To improve upon this approximation, one can introduce an adjustable energy shift  $\delta$  and replace  $\Sigma(E) \rightarrow \Sigma(E - \delta)$  in the effective Hamiltonian. We have determined  $\delta$  empirically, from a fit of theoretical energy levels to the experimental spectrum. Inclusion of this shift mimics high-order corrections in perturbation theory. In addition, we incorporated dressing of the external electromagnetic field (core shielding) in the framework of the random-phase approximation (RPA). To find the valence contribution to  $\alpha_g^{(E1)}(0)$  we summed over the intermediate states in Eq. (3) solving the inhomogeneous equation. A small correction to the polarizability due to core-excited states in Eq. (3) was computed within the relativistic RPA.

The results of calculations for the static electric dipole polarizabilities for the  $ns^2^1S_0$  and  $nsnp^3P_0$  states are presented in Table II. The listed values of the ground-state polarizabilities of Mg, Ca, and Sr were obtained by us earlier [13]. To estimate their uncertainties we used the fact that the intermediate state  $nsnp^1P_1$  contributes to the polarizability at the level of 95–97%. For calculating the polarizabilities we used the best-known literature values of the  $\langle ns^2^1S_0 || D || nsnp^1P_1 \rangle$  matrix elements tabulated in [13]. For Yb  $|\langle 6s^2^1S_0 || D || 6s6p^1P_1 \rangle| = 4.148(2)$  a.u. [16] leading to 0.5% error in  $\alpha_{^1S_0}^1$ . The uncertainties in the polarizabilities of the  $^3P_0$  states were estimated as one-half of the difference between two predictions obtained with  $\delta=0$  and with  $\delta$  determined with the best fit to the experimental energies (thus

mimicking omitted higher-order many-body corrections). The uncertainties in the ground-state polarizabilities range from 0.1% for Sr to 1% for Mg. For the  $^3P_0$  states the errors range from 0.3% for Mg to 6% for Yb. Unlike in the case of the ground state, the polarizability of the  $^3P_0$  states is accumulated due to several transitions: the  $^3D_1$  states contribute only at the level of 50–60%. In Mg the contribution of the  $3s4s\ ^3S_1$  state is even larger than the contribution of the  $3s3d\ ^3D_1$  state. Generally, the accuracy of the calculations becomes worse for heavier atoms; this follows the general trend of many-body calculations, where the correlations, and thus the omitted higher order of perturbation theory, become increasingly important as the number of electrons grows.

With the computed polarizabilities we find the BBR shifts with Eq. (7). The “dynamic” correction  $\eta$  is negligible for the  $^1S_0$  states but is needed for the  $^3P_0$  calculations. Indeed, for the ground state, the smallest excitation energy  $E_{1P_1} - E_{1S_0}$  is equal to  $21\,698\text{ cm}^{-1}$  for Sr. At  $T=300\text{ K}$  the characteristic value of  $y \sim 100$  for all the atoms. By contrast, for the  $^3P_0$  clock level, the transitions to the nearby  $^3D_1$  level involve smaller energies. For Sr, the relevant energy is only  $3841\text{ cm}^{-1}$ , corresponding to a characteristic value of  $y \sim 20$ . At this value, the “static polarizability” approximation  $F_1(y) \approx 4\pi^3/(45y)$  has only a few percent accuracy. While evaluating  $\eta$  we find it sufficient to truncate the summation over intermediate states at the lowest-energy excitation. This “dynamic” correction contributes to the BBR shift of the  $^3P_0$  state at the 0.1% level in Mg, 1% in Ca, 2.7% in Sr, and 0.7% in Yb. Notice that, since the clock BBR shift is

obtained by subtracting BBR shifts of the individual levels, the “dynamic” correction contributes at an enhanced 5% level in Sr. These dynamic corrections must be taken into account if the BBR shifts are derived from dc Stark shift measurements.

Finally, we combine the BBR shifts of the individual clock levels and arrive at the overall BBR corrections summarized in Table I. Our computed BBR shift for Sr,  $-2.354(32)\text{ Hz}$ , is in agreement with the estimate [1] of  $-2.4(1)\text{ Hz}$ . Our uncertainties are better than 3%, except for Yb where the uncertainty is 10%. As discussed in the introduction, although resulting from state-of-the-art relativistic atomic-structure calculations, these uncertainties are still large and substantially affect the projected  $10^{-18}$  fractional accuracy of the lattice-based clocks (see Table I). A potential solution involves operating the clocks at cryogenic temperatures.

At room temperatures, the uncertainties in BBR shifts seem to be a major factor in the error budget of these clocks. At the projected  $10^{-18}$  fractional accuracy, the required accuracies (e.g., 0.02% for Sr) in determining BBR shifts are beyond the presently demonstrated capabilities of atomic calculations and related polarizability measurements.

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