Multipolar theory of black-body radiation shift of atomic energy levels and its implications for optical lattice clocks

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(Dated: January 3, 2022)

A black-body radiation (BBR) shifts of $n s n p ^3P_0 - n s^2 S_0$ clock transition in divalent atoms Mg, Ca, Sr, and Yb are evaluated. A theory of multipolar BBR shifts is developed and its implications are discussed. At room temperatures, the resulting uncertainties in the BBR shifts are relatively large and substantially affect the projected $10^{-18}$ fractional accuracy of the optical-lattice-based clocks.

PACS numbers: 06.30.Ft, 32.10.Dk, 31.25.-v

Atomic clocks based on 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 optical lattice. The lattice laser wavelength is selected in such a way that the dominant perturbation of the clock frequency, the induced ac Stark shifts, for both clock states exactly cancel. Although other effects still perturb the clock frequency, estimates 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 apparent advantage of the optical-lattice clocks has motivated a number of recent proposals: original Katori’s scheme$^2$ with fermionic Sr isotopes has been extended to Mg$^3$, Ca$^4$, and Yb$^5$ atoms and to bosonic isotopes $^8, 9, 10$.

In addition, various schemes of probing the highly-forbidden $n s n p ^3P_0 - n s^2 S_0$ clock transition have been proposed: three-photon transition, electromagnetically-induced transparency, and transition assisted by external magnetic field $7, 8, 9$.

Considering advantages of optical lattice clocks, here we investigate an important systematic effect of the black-body 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. In a laboratory environment with an ambient temperature $T$, one needs to introduce the $T$-dependent BBR correction to the observed frequency. 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 towards the projected $10^{-18}$ accuracy goal; (ii) due to $T^4$ scaling of the BBR shift, it may be beneficial to operate at low temperatures, e.g., at liquid nitrogen 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) 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 general relativistic theory of the BBR shift caused by multipolar (EJ and MJ) components of the radiation field.

| Atom | $\delta \nu_{BBR}$, Hz | $\nu_0$, Hz | $\delta \nu_{BBR}/\nu_0$ | 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   | $-1.25(13)$           | $5.18 \times 10^{14}$ | $-2.4 \times 10^{-15}$ | $3 \times 10^{-16}$ |

The paper is organized as follows. Firstly, we derive relativistic formulae for the BBR shift which incorporate various multipolar contributions of the radiation field. Secondly, we apply these general expressions to determination of the BBR shifts in optical-lattice clocks with Mg, Ca, Sr, and Yb atoms. Unless specified otherwise, atomic units and the Gaussian system of electromagnetic units are used throughout. In these units, $c = 1/\alpha$, where $\alpha = 1/137$ is the fine-structure constant. Temperature is expressed in units of $E_h/k_B$, where $E_h$ is the Hartree energy and $k_B$ is the Boltzmann constant.

*Multipolar theory of the black-body radiation shift.* 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 a difference of the BBR shifts for the two levels. We find that 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 wavenum-
ber of the BBR radiation, 208.51 cm$^{-1}$, at $T = 300$ K, and contributions of the BBR-induced magnetic-dipole and electric-quadrupole transitions to the levels of the manifold may be enhanced. Taking these induced transitions into account requires going beyond the conventional electric-dipole approximation, Ref. [2].

Considering a potential importance of the multipolar contributions, in this section we derive the relevant formulae for BBR-induced energy shifts. Although we develop a general relativistic theory including retardation, at the end of the section we reduce our expressions to practically important non-relativistic non-retarded formulas. The derived expressions are generalization of the electric-dipole BBR shift of Ref. [4].

The BBR spectral density is given by the Plank formula

$$u_\omega (T) = \frac{\alpha^3}{\pi^2} \omega^3 \frac{1}{\exp (\omega/T) - 1}.$$  

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 [3], the BBR shift is given by

$$\delta E_g = \frac{1}{4\alpha^2} \sum_p \int d\hat{k} P.V. \int_0^\infty u_\omega (T) \frac{d\omega}{\omega^2} \alpha^2 \omega \sum_p \left\{ \frac{[\hat{n}_{ap}^+ \hat{n}_{ap}^-]}{\omega_{ap} + \omega} + \frac{[\hat{n}_{ap}^- \hat{n}_{ap}^+]}{\omega_{ap} - \omega} \right\} ,$$  

with averaging over photon propagation directions $\hat{k}$ and polarizations $\alpha$. The second-order summation is over intermediate atomic states $|p\rangle$ and involves the Coulomb-gauge couplings $\hat{\alpha}_j \approx (\alpha \cdot \epsilon) \exp (\frac{\pi}{2} (k \cdot r))$, $\alpha_j$ encapsulating the conventional Dirac matrices. P.V. denotes the Cauchy’s principal value; as elucidated in Ref. [2] it is required for a proper treatment of resonant contributions.

While evaluating matrix elements of operators $\hat{Q}_j$, we use multipolar expansion of $\epsilon (\hat{k} \cdot r)$ in vector spherical harmonics and express the resulting couplings in terms of traditional multipole moments $Q_{JM}^{(0)}$.

$$\langle \hat{n}_{ap}^+ \hat{n}_{ap}^- \rangle = \frac{1}{4\alpha^2} \sum_p \int d\hat{k} P.V. \int_0^\infty u_\omega (T) \frac{d\omega}{\omega^2} \sum_p \left\{ \frac{[\hat{n}_{ap}^+ \hat{n}_{ap}^-]}{\omega_{ap} + \omega} + \frac{[\hat{n}_{ap}^- \hat{n}_{ap}^+]}{\omega_{ap} - \omega} \right\} ,$$  

Here $\lambda = 0$ is for magnetic (MJ) and $\lambda = 1$ is for electric (EJ) multipole amplitudes. Explicit relativistic expressions with retardation for matrix elements of $Q_{JM}^{(0)}$ can be found in Ref. [3]. Neglecting retardation effects (i.e., in the long-wavelength approximation) $Q_{JM}^{(0)}$ become frequency-independent EJ moments $Q_{JM}^{(1)} = r^l C_{JM} (r)$, where $C_{JM} (r)$ are normalized spherical harmonics. In the case of magnetic-dipole transitions in the nonrelativistic limit $Q_{JM}^{(0)} = -\frac{2}{T} (\mathbf{L} + 2\mathbf{S})_M$. Notice that the retardation brings correction in the order of $(\omega T)^2$ to these expressions.

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

$$\delta E_g^{(JM)} = -\frac{\pi J + 1}{J (2J - 1)!!} \alpha^2 \omega \sum_p \int_0^\infty d\omega \omega^{2(J-1)} u_\omega (T) \alpha^2 \omega \sum_p \left\{ \frac{\omega_{pg}}{\omega_{pg} - \omega^2} \right\} ,$$  

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

$$\alpha^{(J)} (\omega) = \frac{2}{2J + 1} \sum_{p,M} | \langle p | Q^{(J)}_M | p \rangle |^2 \left\{ \frac{\omega_{pg}}{\omega_{pg} - \omega^2} \right\} .$$  

A cursory examination of these formulas reveals that compared to $2^J$ multipole, the contribution of $2^{J+1}$ multipole is suppressed by a factor of $\alpha^2 \approx (1/137)^2$. Also for the same $J$ the magnetic contribution is $\alpha^2$ weaker than that of the EJ photons. As in the theory of multipolar radiative transitions $E(J+1)$ and MJ contributions are of the same order in $\alpha$.

To illuminate the $T$-dependence of contributions of individual intermediate states we recast the BBR shifts into a form $\langle J \rangle$ is the total angular momentum of the reference state, $\langle |Q^{(J)}_M| |p\rangle$ is the reduced matrix element

$$\delta E^{(J)} = -\frac{(\alpha T)^{2J+1}}{2J + 1} \sum_p | \langle p | Q^{(J)}_M | p \rangle |^2 F_J \left( \frac{\omega_{pg}}{T} \right) ,$$  

with universal functions

$$F_J (y) = \frac{1}{\pi} \frac{J + 1}{(2J + 1)!! (2J - 1)!!} \sum_p \left\{ \frac{\omega_{pg}}{\omega_{pg} - \omega^2} \right\} \frac{2^{2J+1}}{e^2 - 1} dx .$$  

Functions $F_J (y)$ are multipolar generalizations of function $F_J (y)$ introduced by Farley and Wing [3] 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 change around $y \sim 1$ and slowly fall off for $y \gg 1$. Depending on the value of excitation energy, $\omega_{pg} = y T$, a particular intermediate state may introduce either negative or positive BBR shift. Notice that $F_J$ are broad distributions, 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

\[
\delta E^{(j\lambda)}_g = -\frac{\zeta(2J+2)(2J+1)!}{2\pi J [(2J-1)!]^2} \alpha^{2J+1} T^{2J+2} \alpha_{g}^{(J\lambda)}(0),
\]

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 multipolarity \( J \) increases by one, 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 \( (k_B T / E_h)^2 \approx 9.0 \times 10^{-7} \).

BBR shift for \( ^3P_0 - ^1S_0 \) transition in divalent atoms.

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 \( \delta \nu_{BBR} \) is the difference between the two individual shifts, \( \delta \nu_{BBR} = \delta \nu_{BBR}(^3P_0) - \delta \nu_{BBR}(^1S_0) \).

Consider first the BBR shift of the ground \( ^1S_0 \) state. Here transition energies of various multipole 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 would 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 \( 5 \times 10^{-15} \) level (see Table 1), 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 \( \delta E^{(M1)}_g \sim \alpha^2 (\alpha T)^3, \delta E^{(E2)}_g \sim (\alpha T)^5 \), while \( \delta E^{(E1)}_g \sim \alpha^3 (T)^4 / \omega_{P_2 - P_0} \). Our numerical estimate, based on the transitions inside the fine-structure manifold lead to the following values of the BBR shifts for Sr: \( \delta E^{(M1)}_g = 2.4 \times 10^{-5} \) Hz and \( \delta E^{(E2)}_g = 2.5 \times 10^{-8} \) 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 universal functions \( F_J \) have comparable values for a wide range of excitation energies, \( |\omega| \lesssim 20 \) T, see Fig. 1. For example, for Sr the \( ^3P_0 - ^3D_1 \) 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. 11 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,

\[
\delta E^{(E1)}_g \approx -\frac{2}{15} (\alpha \pi)^3 T^4 \alpha_{g}^{(E1)}(0) \times [1 + \eta],
\]

\[
\eta = \frac{y_p (80/63)^{5} \pi^{2}}{\alpha_{g}^{(E1)}(0)} \sum_{p} \frac{|\langle g | Q^{(1)}_{1} | y \rangle|^{2}}{(2J_1 + 1)y_p^3} \left( 1 + \frac{21 \pi^2}{5y_p} + \frac{336 \pi^4}{11y_p^3} \right).
\]

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 asymptotic expansion \( F_1(y) \approx \frac{4\pi}{45} + \frac{32\pi}{189y_p} + \frac{32\pi^7}{45y_p^3} + \frac{512\pi^9}{99y_p^5} \), which has an accuracy better than 0.1% for \( |y| \gg 1 \). \( \eta \) 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 methods of atomic structure.

Evaluation of the static dipole polarizabilities follows a procedure of Ref. 11. Here we only outline the major steps. We employ relativistic many-body code described in Refs. 12, 13, 14. The employed formalism is a combination of configuration-interaction method in the valence space with many-body perturbation theory for core-polarization effects. In this method, one determines wave functions from solving the effective many-body Schrödinger equation

\[
\{ H_{FC} + \Sigma(E) \} |\Psi_n\rangle = E_n |\Psi_n\rangle,
\]

Here \( H_{FC} \) 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 \( \Sigma(E) \to \Sigma(E - \delta) \) in the effective Hamiltonian, Eq. 8. We have determined \( \delta \) empirically, from a fit of theoretical energy levels to 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
TABLE II: Static electric dipole polarizabilities in a.u. and BBR shifts in Hz for the ground $^3S_0$ and the lowest-energy $^3P_0$ excited states of Mg, Ca, Sr, and Yb atoms. Theoretical uncertainties are indicated in parenthesis.

|          | Mg       | Ca       | Sr       | Yb       |
|----------|----------|----------|----------|----------|
| $\alpha_{P_0}^{(E1)}$ | 101.2(3) | 290.3(15) | -0.872(3) | -2.29(13) |
| $\delta \alpha_{P_0}^{(E1)}$ | -0.614(6) | -1.353(11) | -1.698(2) | -1.04(3) |

valence contribution to $\alpha_{g}^{(E1)}(0)$ we summed over the intermediate states in Eq. 8 using the Dalgaro-Lewis-Sternheimer method 17. A small correction to polarizability due to core-excited intermediate states in Eq. 8 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 11. To estimate their uncertainties we used the fact that the intermediate state polarizabilities were estimated as a half of the difference $\delta \alpha_{P_0}^{(E1)}$. The uncertainties in the ground-state polarizabilities range from 0.1% for Sr to 3% for Yb. For the $^3P_0$ states the errors range from 0.3% for Mg to 6% for Yb.

With the computed polarizabilities we can find the BBR frequency 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 $E_{1P_0} - E_{1S_0}$ is equal to 21698 cm$^{-1}$ for Sr. At $T = 300 K$ 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 cm$^{-1}$ corresponding to characteristic value of $y \sim 20$. At this value, the “static polarizability” approximation, $F_1(y) \approx 4\pi^2/(45y)$, 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 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 III. Our computed BBR shift for Sr, $-2.354(32)$ Hz is in agreement with an estimate 18 of $-2.4(1)$ Hz. Our uncertainties are better than 3%, except for Yb where the error is 10%. As discussed in the introduction, although resulting from state-of-the-art relativistic atomic-structure calculations, these errors are still large and substantially affect the projected $10^{-18}$ fractional accuracy of the lattice-based clocks (see Table I). Potential solution involves operating the clocks at cryogenic temperatures.

We would like to thank C. Oates and E. N. Fortson for motivating discussions. This work was supported in part by the NSF Grant No. PHY-0354876; NIST precision measurement grant, and RFBR Grant Nos. 04-02-16345-a and 05-02-16914-a.

[1] M. Takamoto, F. L. Hong, R. Higashi, and H. Katori, Nature 435, 321 (2005).
[2] H. Katori, in Proc. 6th Symposium Frequency Standards and Metrology, edited by P. Gill (World Scientific, Singapore, 2002), pp. 323–330.
[3] Jan W. Thomsen, private communication.
[4] F. Richle et al., talk at the Second Workshop on Cold Alkali-Earth Atoms, September 11-13, 2003, Copenhagen, Denmark.
[5] S. G. Porsev, A. Derevianko, and E. N. Fortson, Phys. Rev. A 69, 021403(R) (2004).
[6] R. Santra, E. Arimondo, T. Ido, C. H. Greene, and J. Ye, Phys. Rev. Lett. 94, 173002 (2005).
[7] T. Hong, C. Cramer, W. Nagourney, and E. N. Fortson, Phy. Rev. Lett. 94, 050801 (2005).
[8] A. V. Talichenachev, V. I. Yudin, C. W. Oates, C. W. Hoyt, Z. W. Barber, and L. Hollberg (2005), e-print arXive: physics/0510048
[9] J. W. Farley and W. H. Wing, Phys. Rev. A 23, 2397 (1981).
[10] W. R. Johnson, D. R. Plante, and J. Sapirstein, Adv. At. Mol. Phys. 35, 255 (1995).
[11] S. G. Porsev, A. Derevianko, Zh. Eksp. Teor. Fiz. 129, 227 (2006), [Sov. Phys.-JETP 102 195, (2006)].
[12] V. A. Dzuba, V. V. Flambaum, and M. G. Kozlov, Phys. Rev. A 54, 3948 (1996).
[13] V. A. Dzuba, M. G. Kozlov, S. G. Porsey, and V. V. Flambaum, Zh. Eksp. Teor. Fiz. 114, 1636 (1998), [Sov. Phys.-JETP 84 461, (1997)].
[14] M. G. Kozlov and S. G. Porsey, Eur. Phys. J. D 5, 59 (1999).
[15] A. Dalgarno and J. T. Lewis, Proc. Roy. Soc. 223, 70 (1955).