Blackbody radiation shift for the $^{1}S_{0} \rightarrow ^{3}P_{0}$ optical clock transition in zinc and cadmium atoms

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Abstract
Black-body radiation (BBR) shifts of the $^{1}S_{0} \rightarrow ^{3}P_{0}$ clock transition in divalent atoms Cd and Zn are evaluated using accurate relativistic many-body techniques of atomic structure. Static polarizabilities of the clock levels and relevant electric-dipole matrix elements are computed. We also present a comparative overview of the BBR shifts in optical clocks based on neutral divalent atoms trapped in optical lattices. Zinc and cadmium atoms have one of the smallest BBR clock shifts, mitigating the largest inaccuracy in optical lattice clocks.

Keywords: atomic clock, black body radiation, $ab$ initio atomic structure calculations

One of the factors limiting the accuracy of the modern atomic clocks is the perturbation of the clock frequency by the bath of thermal photons, i.e. by black body radiation (BBR). $10^{-15}$ is the typical value of the fractional BBR correction to optical lattice clocks [1] at room temperatures, while the current generation of optical atomic clocks have demonstrated the fractional inaccuracies at the level of $10^{-18}$ or better [2–4]. Therefore, all the recent advances in atomic clocks address the BBR shift problem either through cryogenic techniques, active temperature stabilization, or specially-designed BBR chambers. All of these techniques can be advanced further by using atoms that have a reduced sensitivity to BBR.

To the leading order, the fractional BBR correction to the unperturbed clock frequency $\nu_0$ can be parameterized as

$$\frac{\delta \nu}{\nu_0} = \beta \left( \frac{T}{300 \text{ K}} \right)^4,$$

where $T$ is the bath temperature. There are two issues associated with the BBR shift: (i) one needs to know the coefficient $\beta$ with sufficiently high-accuracy so that the uncertainty in $\beta$ does not degrade the clock output and (ii) even if $\beta$ is known precisely, there are uncertainties arising from the ambient temperature fluctuations and imperfect knowledge of the temperature field. Apparently, the smaller the $\beta$, the better.

There are two main classes of optical atomic clocks that are presently well-positioned to eventually replace the primary frequency standard. The first, more mature, class of clocks is based on trapped ions and the second class employs neutral divalent atoms trapped in optical lattices. A comparative overview of the BBR shift for various ion clocks is given in [5] and for lattice clocks in [1]. The NIST group [6] has pointed out that the BBR shift is exceptionally small in Al$^+$ ion, $\delta \nu/\nu_0 \sim 10^{-17}$. For divalent atoms considered in the literature so far (Mg, Ca, Sr, Yb, Hg) the least susceptible are mercury lattice clocks [7], $\delta \nu/\nu_0 \sim 10^{-16}$ at room temperatures.

Divalent cadmium and zinc atoms were found recently [8] to have properties suitable for realizing the neutral atom optical lattice clocks. With the BBR shift being one of the most important contributors to the uncertainty budget of the clocks, here we extend the survey of [1] and compute the BBR shifts for the Cd and Zn lattice clocks. The results of our analysis are summarized in table 1. We find that for Cd and Zn the fractional BBR shifts are comparable to the so-far most favorable Hg. At least from this perspective, these atoms may serve as a competitive alternative to already operational Sr, Yb, and Hg clocks.
| Atom | \(\ell\nu_0\) (Hz) | \(\nu_0\) (Hz) | \(\delta\nu_0/\nu_0\) | Uncertainty |
|------|-----------------|-----------------|----------------------|-------------|
| Zn   | -0.244(10)      | 9.69 \times 10^{14} | -2.5 \times 10^{-16} | 1 \times 10^{-17} |
| Cd   | -0.248(15)      | 9.03 \times 10^{14} | -2.8 \times 10^{-16} | 2 \times 10^{-17} |
| Mg [1] | -0.258(7)      | 6.55 \times 10^{14} | -3.9 \times 10^{-16} | 1 \times 10^{-17} |
| Ca [1] | -1.171(17)      | 4.54 \times 10^{14} | -2.6 \times 10^{-15} | 4 \times 10^{-17} |
| Sr [1] | -2.354(32)      | 4.29 \times 10^{14} | -5.5 \times 10^{-15} | 7 \times 10^{-17} |
| Yb [1] | -1.25(13)       | 5.18 \times 10^{14} | -2.4 \times 10^{-15} | 3 \times 10^{-16} |
| Hg [7] | -0.181         | 1.13 \times 10^{15} | -1.6 \times 10^{-16} |             |

### Details of calculations

To compute the energy shift due to black body radiation we use the formalism developed in [1]. The electric-dipole contribution to the BBR energy shift of state \(v\) is given by

\[
\delta E_v \approx -\frac{2}{15} (\alpha\pi)^2 T^3 \alpha_s(0) [1 + \eta].
\]

\[
\eta = \frac{80/63\pi^2}{\alpha_s(0) T} \sum_p \left( \frac{|p|!|D||n|!}{(2J_p + 1)y_p^2} \right) \left( 1 + \frac{21\pi^2}{5y_p^2} + \frac{336\pi^4}{111y_p^4} \right).
\]

(1)

Here \(y_p = (E_p - E_0)/T\), \(\alpha_s(0)\) is the static scalar dipole polarizability, and \(\eta\) represents a ‘dynamic’ fractional correction to the total shift. \(D\) is the electric-dipole operator. The calculations require evaluating the static polarization for both clock levels. The clock transition is between the \(^1S_0\) ground state and the lowest-energy \(^3P_0\) state.

The static scalar polarizability \(\alpha_s(0)\) of an atom in state \(v\) is given by

\[
\alpha_s(0) = \frac{2}{3(2J_v + 1)} \sum_n \frac{|v||D||n|!^2}{E_n - E_v}. \tag{2}
\]

where summation goes over the complete set of excited many-body states (including continuum and core-excited states). We use the Dalgarno–Lewis method and reduce the summation to solving the inhomogeneous Schrödinger (Dirac) equation (setup is similar to [11]). In this approach, a correction to the atomic wave function due to the external electric field is introduced

\[
\langle \delta \Psi_v | = \sum_n \frac{|v||D||n|}{E_n - E_v} (\eta). \tag{3}
\]

This correction satisfies an inhomogeneous equation

\[
(\tilde{H}_0 - E_v) \delta \Psi_v = -D \Psi_v, \tag{4}
\]

where \(\tilde{H}_0\) is an effective Hamiltonian of the atom. Once the \(\delta \Psi_v\) is found, static polarizability is calculated as

\[
\alpha_s(0) = \frac{2}{3(2J_v + 1)} \langle \delta \Psi_v | D | v \rangle. \tag{5}
\]

We employ a computational scheme based on combining the configuration-interaction method with the many-body perturbation theory (CI+MBPT) [12]. The effective Hamiltonian is constructed for the two valence electrons, while excitations from the core are taken into account by means of the MBPT. The Hamiltonian has the form

\[
\tilde{H}_0 = \hat{h}_1(1) + \hat{h}_1(2) + \hat{h}_{12}, \tag{6}
\]

where \(\hat{h}_1\) is a single-electron part of the relativistic Hamiltonian

\[
\hat{h}_1 = c(c \cdot p) + mc^2\ell(\ell - 1) - \frac{Ze^2}{r} + \hat{V}_{\text{core}} + \hat{\Sigma}_1, \tag{7}
\]

Here \(c\) is speed of light, and \(\alpha\) and \(\beta\) are Dirac matrices, \(Ze\) is the nuclear charge, \(\hat{V}_{\text{core}}\) is the Hartree–Fock potential of the atomic core (including the non-local exchange term) and \(\hat{\Sigma}_1\) is the correlation potential which describes the interaction between a valence electron and the core (see [12, 13] for details).

The \(\hat{h}_{12}\) operator in (6) is the two-electron part of the Hamiltonian:

\[
\hat{h}_{12} = \frac{\hat{r}^2}{\ell_{12}^2} + \hat{\Sigma}_2, \tag{8}
\]

where first term is standard Coulomb interaction between valence electrons and second term is the correction to it due to correlations with core electrons.

We use the second-order MBPT to calculate the self-energy operators \(\hat{\Sigma}_1\) and \(\hat{\Sigma}_2\) via direct summation over a complete set of single-electron states. This set of basis states is constructed using the B-spline technique [14]. We use 40 B-splines of order 9 in a cavity of 40 Bohr radius. The same basis of the single-electron states is also used in constructing the two-electron basis states for the CI calculations. We employ partial waves \(\ell = 0 - 4\) for the valence CI subspace and \(\ell = 0 - 5\) for internal summations inside the self-energy operator.

Additionally, to mimic the omitted higher-order MBPT effects, we rescale the \(\hat{\Sigma}\) operator to fit the experimental energies. The \(\hat{\Sigma}_1\) operator is replaced in (6) by \(\lambda \hat{\Sigma}_1\), where \(l = 0, 1, 2\) is the angular momentum of a single-electron state. The \(\hat{\Sigma}_2\) operator is replaced by \(f_k \hat{\Sigma}_2\), where \(k\) is

\[
\lambda = \frac{1}{9} \left( \frac{1003}{1000} \right)^2, f_k = \left( \frac{\ell + 1/2}{8} \right)^2. \tag{9}
\]

\[
\nu_0 = \frac{\delta\nu_0}{\nu_0}. \tag{10}
\]

\[
\ell_{12} = \frac{2}{3} \left( \frac{\ell + 1}{\ell} \right)^2. \tag{11}
\]

\[
\hat{D} = \frac{1}{2} \left( \frac{2\ell + 1}{\ell} \right)^2. \tag{12}
\]
multipolarity of the Coulomb interaction. The values of the rescaling parameters are presented in table 2. The resulting energies after the scaling procedure are listed in table 3. A typical deviation from the experimental values is in the order of 100 cm$^{-1}$. Even after the scaling, the disagreement remains, as the number of fitting parameters is limited.

### Results

With the computed wavefunctions, we may evaluate various matrix elements. While computing matrix elements (and polarizabilities) we use single-particle matrix elements dressed in the random-phase approximation. Qualitatively this corresponds to the shielding of the applied electromagnetic field by the core electrons. Notice that the static polarizability depends sensitively on the values of the dipole matrix elements for the lowest-energy excitations. Our computed dipole matrix elements for the two lowest-energy excitations originating from the two clock states are presented in table 4. The inter-combination transition $^1S_0 \rightarrow ^3P_1^o$ is non-relativistically forbidden. A three-fold increase in the matrix element values when progressing from Zn $Z = 30$ to heavier Cd $Z = 48$ is consistent with the relevant suppression factor of $(\alpha Z)^2$.

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