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Blackbody radiation shift for the $^1S_0 - ^3P_0$ optical clock transition in zinc and cadmium atoms

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Abstract

Black-body radiation (BBR) shifts of the $^1S_0 - ^3P_0$ 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 ν_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 β with sufficiently high-accuracy so that the uncertainty in β does not degrade the clock output and (ii) even if β is known precisely, there are uncertainties arising from the

ambient temperature fluctuations and imperfect knowledge of the temperature field. Apparently, the smaller the β , 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.

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Table 1. Black-body radiation shift for the clock transitions between the 1S_0 ground state and the lowest-energy $^3P_0^o$ state for divalent atoms. Result for Zn and Cd are from this work, for Mg, Ca, Sr, Yb from [1], and for Hg from [7]. $\delta\nu$ is the BBR shift at $T = 300$ K with our estimated uncertainties. ν_0 is the clock transition frequency, and $\delta\nu/\nu_0$ 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. These uncertainties are derived from theoretical estimates. More recent experimental work for Sr and Yb (measuring polarizabilities, state lifetimes, etc) has constrained the fractional BBR shift uncertainties to the 10^{-18} level at room temperature [9, 10].

Atom	$\delta\nu$, (Hz)	ν_0 , (Hz)	$\delta\nu/\nu_0$	Uncertainty
Zn	-0.244(10)	9.69×10^{14}	-2.5×10^{-16}	1×10^{-17}
Cd	-0.248(15)	9.03×10^{14}	-2.8×10^{-16}	2×10^{-17}
Mg [1]	-0.258(7)	6.55×10^{14}	-3.9×10^{-16}	1×10^{-17}
Ca [1]	-1.171(17)	4.54×10^{14}	-2.6×10^{-15}	4×10^{-17}
Sr [1]	-2.354(32)	4.29×10^{14}	-5.5×10^{-15}	7×10^{-17}
Yb [1]	-1.25(13)	5.18×10^{14}	-2.4×10^{-15}	3×10^{-16}
Hg [7]	-0.181	1.13×10^{15}	-1.6×10^{-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 ν is given by

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

$$\eta = \frac{(80/63)\pi^2}{\alpha_\nu(0)T} \sum_p \frac{|\langle p || D || \nu \rangle|^2}{(2J_\nu + 1)y_p^3} \left(1 + \frac{21\pi^2}{5y_p^2} + \frac{336\pi^4}{11y_p^4} \right). \quad (1)$$

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

The static scalar polarizability $\alpha_\nu(0)$ of an atom in state ν is given by

$$\alpha_\nu(0) = \frac{2}{3(2J_\nu + 1)} \sum_n \frac{|\langle \nu || \mathbf{D} || n \rangle|^2}{E_n - E_\nu}, \quad (2)$$

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

$$\langle \delta \Psi_\nu | = \sum_n \frac{\langle \nu || \mathbf{D} || n \rangle}{E_n - E_\nu} \langle n |. \quad (3)$$

This correction satisfies an inhomogeneous equation

$$(\hat{H}_0 - E_\nu) \delta \Psi_\nu = -\mathbf{D} \Psi_\nu, \quad (4)$$

where \hat{H}_0 is an effective Hamiltonian of the atom. Once the $\delta \Psi_\nu$ is found, static polarizability is calculated as

$$\alpha_\nu(0) = \frac{2}{3(2J_\nu + 1)} \langle \delta \Psi_\nu || \mathbf{D} || \nu \rangle. \quad (5)$$

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

$$\hat{H}_0 = \hat{h}_1(1) + \hat{h}_1(2) + \hat{h}_{12}, \quad (6)$$

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

$$\hat{h}_1 = c(\boldsymbol{\alpha} \cdot \mathbf{p}) + mc^2(\beta - 1) - \frac{Ze^2}{r} + \hat{V}_{\text{core}} + \hat{\Sigma}_1. \quad (7)$$

Here c is speed of light, and $\boldsymbol{\alpha}$ and β are Dirac matrices, Ze is the nuclear charge, \hat{V}_{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 correlation 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:

$$\hat{h}_{12} = \frac{e^2}{r_{12}} + \hat{\Sigma}_2, \quad (8)$$

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 $\lambda_l \hat{\Sigma}_1$, where $l = 0, 1, 2$ is a the angular momentum of a single-electron state. The $\hat{\Sigma}_2$ operator is replaced by $f_k \hat{\Sigma}_2$, where k is

Table 2. Rescaling parameters λ_l and f_k for the core-valence correlation operators $\hat{\Sigma}_1$ and $\hat{\Sigma}_2$.

	λ_s	λ_p	f_1
Zn	1.113	1.106	1
Cd	0.871 4	0.887	0.8

Table 3. Energy levels of Zn and Cd (cm^{-1}); comparison of ‘scaled’ theory and experiment. $n = 4$ for Zn and $n = 5$ for Cd.

Config.	State	J	Zn		Cd	
			Expt.	Theory	Expt.	Theory
<i>nsnp</i>	$^3P^o$	0	32 311	32 348	30 114	30 108
		1	32 501	32 546	30 656	30 664
		2	32 890	32 950	31 827	31 866
<i>nsnp</i>	$^1P^o$	1	46 745	46 908	43 692	43 721
<i>ns(n + 1)s</i>	3S	1	53 672	53 412	51 484	51 317
<i>ns(n + 1)s</i>	1S	0	55 789	55 513	53 310	53 088
<i>nsnd</i>	1D	2	62 459	62 333	59 220	59 282
<i>nsnd</i>	3D	1	62 769	62 606	59 486	59 512
		2	62 772	62 609	59 498	59 521
		3	62 777	62 613	59 516	59 534

Table 4. Electric dipole transition amplitudes (reduced matrix elements, a.u.) for Zn and Cd.

Transition	Zn	Cd
$^1S_0 \rightarrow ^3P_1^o$	0.045	0.158
$^1S_0 \rightarrow ^1P_1^o$	3.320	3.435
$^3P_0^o \rightarrow ^3S_1$	1.466	1.486
$^3P_0^o \rightarrow ^3D_1$	2.127	2.222

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 $(\alpha Z)^2$.

Table 5. Static polarizabilities of the 1S_0 and $^3P_0^o$ states of Zn and Cd (a.u.); comparison with experimental results and other calculations.

Atom	State	Expt. [18, 19]	This work	[20]	[21]
Zn	1S_0	38.8(8)	38.58	38.12	39.13
Zn	$^3P_0^o$		66.53	67.69	66.50
Cd	1S_0	49.65(1.62)	46.52	44.63	
Cd	$^3P_0^o$		75.31	75.29	

Similarly to the case of Sr and Yb atoms [15, 16], we anticipate that the high-accuracy values for the $^1S_0 \rightarrow ^1P_1^o$ matrix elements may be derived from photoassociation spectroscopy with ultracold atoms. If such data become available, the accuracy of our values for polarizability may be improved by correcting the matrix elements of table 3 with the experimental values and correcting $\alpha_v(0)$ with Equation (2).

The computed values of the static polarizabilities of the clock levels are presented in table 5. The values combine both valence and core polarizabilities. Core polarizabilities are 2.296 a.u. for Zn and 4.971 a.u. for Cd [17]. For the ground states we compare our values with the experimental results [18, 19]. For Zn our computed value is within the experimental uncertainty while for Cd the results disagree by about 2σ of the experiment. Our results are consistent with the previous theoretical work [20, 21]. These authors employed methods sufficiently different from our approach to warrant additional confidence in the theoretical predictions. [20] used a semi-empirical model potential method and [21] employed a multi-reference configuration-interaction method using a two-electron relativistic pseudo-potential.

Finally, we combine the static polarizabilities using equation (1) and arrive at the BBR shifts summarized in table 1. The results also include the dynamic correction η ; it turns out to be less than 7×10^{-4} for both Zn and Cd. This small correction can be safely neglected at the present level of accuracy. We also estimate the theoretical error bar for the BBR correction: 4% for Zn and 6% for heavier Cd. The error was evaluated by carrying out two calculations: with and without scaling of self-energy operator to experimental energies. We find that the resulting uncertainty would affect the accuracy of the clock output in the 17th significant figure. Overall fractional BBR shifts for both Cd and Zn are slightly larger than in Hg, but 5 times smaller than in Sr and 10 times smaller than in Yb.

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