

## Role of the multipolar black-body radiation shifts in the atomic clocks at the $10^{-18}$ uncertainty level

B K SAHOO

Theoretical Physics Division, Physical Research Laboratory, Ahmedabad 380 009, India  
E-mail: bijaya@prl.res.in

DOI: 10.1007/s12043-014-0791-9; ePublication: 27 July 2014

**Abstract.** We present here an overview of the role of the multipolar black-body radiation (BBR) shifts in the single ion atomic clocks to appraise the anticipated  $10^{-18}$  uncertainty level. With an attempt to use the advanced technologies for reducing the instrumental uncertainties at the unprecedented low, it is essential to investigate contributions from the higher-order systematics to achieve the ambitious goal of securing the most precise clock frequency standard. In this context, we have analysed contributions to the BBR shifts from the multipolar polarizabilities in a few ion clocks.

**Keywords.** Atomic clock; polarizability; spectroscopy; many-body theory.

**PACS Nos** 31.15.bw; 32.10.Dk; 06.30.Ft; 32.80.–t

### 1. Introduction

With the advent of many sophisticated technologies, it is possible today to measure atomic transition frequencies more precisely than yesteryears. Due to the achievement of high quality factor  $Q$ , the ratio between the uncertainty and the absolute frequency of an atomic clock, it is now under debate to replace the present primary Cs atomic clock with one of the recent time clocks with high stability, reproducibility and accuracy; in other words, the considered frequency standard can be reproducible over time and space within the reported uncertainty. The possible promising candidates that are capable of replacing the primary Cs atomic clock are: neutral atoms trapped in the optical lattices and/or the trapped singly positive ions, the advantages of which are their stability due to their confinement in a strong electric field because of the advanced trap techniques with optical frequency combs that locks the laser frequencies more precisely.

Here, we shall focus our discussion only on the atomic clocks using the singly charged ions whose clock frequencies are in the optical region. The most promising ions that are under consideration for the purpose of making frequency standards are  $\text{Hg}^+$  [1],  $\text{Ca}^+$  [2],  $\text{Sr}^+$  [3],  $\text{Al}^+$  [4] and  $\text{Yb}^+$  [5]. Among these, the fractional uncertainties in  $\text{Hg}^+$  and  $\text{Al}^+$  have already reached  $10^{-17}$  level [6]. A few more ions such as  $\text{Ba}^+$  [7,8],  $\text{Ra}^+$  [9,10]

etc. are also proposed. These clocks will help us not only to define the SI second more precisely, but also can be used for making the global positioning systems, tracking of deep-space probes, inferring fundamental physics like probing possible variation of the fine-structure constant etc.

In most of these ions mentioned, the main issue is to reduce the systematic effects and/or their uncertainties so that they can be considered as the most precise atomic clocks. In that respect, the black-body radiation (BBR) shift is one of the significant systematic effects in many of the atomic ions considered for the clock purposes and to precisely estimate its shift is a matter of current interest. Precise measurements of this effect would require sophisticated instruments, it may be costly and may need huge manpower. In contrast, any accurate relativistic many-body approaches relying on the high performance computing systems that are capable of describing the atomic structures correctly can also be used to estimate the BBR shifts with the desired precision but at a lower cost. As the estimate of BBR shifts in a particular atomic clock depends directly on the differential scalar polarizability of the associated states in the clock transition, this quantity in an atomic system can be calculated accurately; a number of accurate calculations are reported (e.g., [9,11–14] and references therein). However, most of the calculated BBR shifts are determined by accounting contributions only from the electric dipole ( $E1$ ) polarizabilities assuming that contributions from the other polarizabilities are negligible or lower than the required precision. The considered transitions in the singly charged alkaline earth atomic ions are basically the  $S_{1/2} \rightarrow D_{3/2}$  and/or  $S_{1/2} \rightarrow D_{5/2}$  transitions. Typically, the magnetic dipole ( $M1$ ) polarizabilities of the  $S_{1/2}$  states are small in magnitudes, but these quantities are large for the  $D_{3/2}/D_{5/2}$  states due to the large contributions from their counter fine structure levels. In an attempt to push the relative uncertainties in these ion clocks below  $10^{-17}$  level, we verified recently the contributions from the multipolar polarizabilities in two prominent ion clocks,  $\text{Ca}^+$  and  $\text{Sr}^+$  [14], and found that the contributions from these higher multipolar polarizabilities are significant at the  $10^{-18}$  level. In this paper, we are extending similar studies to find out contributions from the multipolar polarizabilities to the BBR shifts in the proposed  $S_{1/2} \rightarrow D_{5/2}$  clock transitions in  $\text{Ba}^+$  and  $\text{Ra}^+$  to verify their roles at the  $10^{-18}$  uncertainty level.

## 2. Theory

In the presence of a weak electromagnetic field, the energy shift of an atomic energy level  $|\Psi_n\rangle$  with energy  $E_n (= \omega_n)$  is approximated to [14–16]

$$\delta E_n(\omega) = \frac{1}{2} \sum_{m \neq n} |V_{nm}(r, \omega)|^2 \left( \frac{\omega_n - \omega_m}{(\omega_n - \omega_m)^2 - \omega^2} \right), \quad (1)$$

where the external propagating electromagnetic field in the Coulomb gauge is given by

$$V(r, \omega) = -c \boldsymbol{\alpha} \cdot \mathbf{A}(r, \omega) = -c (\boldsymbol{\alpha} \cdot \hat{\boldsymbol{\epsilon}}) \exp(i\mathbf{k} \cdot \mathbf{r}) \quad (2)$$

with  $\alpha$  being the Dirac matrix,  $\omega$  is the angular frequency of the field and  $\mathbf{k} = k\hat{k}$  and  $\hat{\epsilon}$  are its wave vector and polarization direction, respectively.

With the relations  $\mathbf{B} = \nabla \times \mathbf{A}$  and  $\mathbf{E} = (i\omega/c)\mathbf{A}$ , the energy shift after averaging over  $\omega$  for all the polarization and propagation directions is given as [16]

$$\delta E_n^{(\lambda,L)} = -\frac{(\alpha k_B T)^{2L+1}}{2J_n + 1} \sum_{m \neq n} |\langle \Psi_n \| Q_L^\lambda \| \Psi_m \rangle|^2 F_L \left( \frac{\omega_{mn}}{k_B T} \right), \quad (3)$$

where  $Q_L^\lambda$  are the multipolar electromagnetic transition operators due to  $V(r, \omega)$ ,  $J_n$  is the angular momentum of state  $|\Psi_n\rangle$ ,  $\lambda = 1$  and  $\lambda = 0$  correspond to the electric and magnetic multipoles, respectively, and

$$F_L(y) = \frac{1}{\pi} \frac{L+1}{L(2L+1)!!(2L-1)!!} \int_0^\infty \left( \frac{1}{y+x} + \frac{1}{y-x} \right) \frac{x^{2L+1}}{e^x - 1} dx. \quad (4)$$

Here, the argument  $y = \omega_{mn}/(k_B T) = (\omega_n - \omega_m)/(k_B T)$ . The function  $F_L(y)$  is a universal function applicable to all the atoms with argument  $y$  depending on the range of the atomic parameters; the valid limit for these functions in our case is  $|y| \gg 1$ .

Using the general definition of the scalar polarizability,

$$\alpha_n^{L,\lambda} = \frac{2}{\alpha^{2(\lambda-1)}(2L+1)(2J_n+1)} \sum_{m \neq n} \frac{|\langle \Psi_n \| Q_L^\lambda \| \Psi_m \rangle|^2}{E_n - E_m}, \quad (5)$$

the BBR shift for different channels are given by

$$\delta E_n^{E1} = -\frac{1}{2} \frac{4\pi^3 \alpha^3}{15} (k_B T)^4 \alpha_n^{E1} = -\frac{1}{2} \langle E_{E1}^2(\omega) \rangle \alpha_n^{E1}, \quad (6)$$

$$\delta E_n^{M1} = -\frac{1}{2} \frac{4\pi^3 \alpha^5}{15} (k_B T)^4 \alpha_n^{M1} = -\frac{1}{2} \alpha^2 \langle B_{M1}^2(\omega) \rangle \alpha_n^{M1} \quad (7)$$

and

$$\delta E_n^{E2} = -\frac{1}{2} \frac{8(\alpha\pi)^5 (k_B T)^6}{189(2J_n+1)} \alpha_n^{E2} = -\frac{1}{2} \langle E_{E2}^2(\omega) \rangle \alpha_n^{E2}, \quad (8)$$

with  $\alpha_n^{E1}$ ,  $\alpha_n^{M1}$  and  $\alpha_n^{E2}$  being the scalar  $E1$ ,  $M1$  and  $E2$  polarizabilities, respectively. Here,  $E_{E1}$ ,  $B_{M1}$  and  $E_{E2}$  are the electric and magnetic field densities that are defined as

$$E_L(\omega) = B_L(\omega) = \sqrt{\frac{8}{3\pi c^3} \frac{\omega^3}{\exp(\omega/k_B T) - 1}}. \quad (9)$$

Substituting all, the BBR shifts (in Hz) with respect to the room temperature yield [14]

$$\delta E_{f \rightarrow i}^{E1} = \delta E_f^{E1} - \delta E_i^{E1} = -\frac{1}{2} (831.9 \text{ V/m})^2 \left[ \frac{T(\text{K})}{300} \right]^4 (\alpha_f^{E1} - \alpha_i^{E1}), \quad (10)$$

$$\begin{aligned} \delta E_{f \rightarrow i}^{M1} &= \delta E_f^{M1} - \delta E_i^{M1} \\ &= -\frac{1}{2} (2.77 \times 10^{-6} \text{ T})^2 \left[ \frac{T(\text{K})}{300} \right]^4 (\alpha_f^{M1} - \alpha_i^{M1}) \end{aligned} \quad (11)$$

and

$$\begin{aligned}\delta E_{f \rightarrow i}^{E2} &= \delta E_f^{E2} - \delta E_i^{E2} \\ &= -\frac{1}{2}(7.2 \times 10^{-3} \text{V/m})^2 \left[ \frac{T(\text{K})}{300} \right]^6 (\alpha_f^{E2} - \alpha_i^{E2}).\end{aligned}\quad (12)$$

### 3. Method of calculation

We use the relativistic coupled-cluster (RCC) approach to determine the atomic wave functions in the considered systems. As the configurations of the atomic states involved in the clock transitions have a closed-core structure as with a valence electron, we adopt a two-step procedure to calculate the wave functions for these states. In a Fock-space approach, we first calculate the Dirac–Fock (DF) wave function with the closed-shell ( $|\Phi_0\rangle$ ), then append the corresponding valence orbital  $v$  to define DF or the reference state ( $|\Phi_v\rangle = a_v^\dagger |\Phi_0\rangle$ ) for the desired atomic state. The correlation effects among these orbitals are then accounted for in three forms: core, core-valence and valence correlation effects. As valence orbitals are a part of the virtual space in our approach, the core and core-valence correlations are considered by the excitation operators ( $T$ ) by exciting electrons from the occupied orbitals to virtual and valence orbitals, respectively. The valence correlation effects are considered by other excitation operators ( $S_v$ ) that excite electrons from either the valence orbital alone or along with core orbitals. In the RCC framework, these atomic wave functions can be put in a mathematical form as [8,9,11,12]

$$|\Psi_v\rangle = e^T \{1 + S_v\} |\Phi_v\rangle. \quad (13)$$

The amplitudes for various excitations are solved first for the  $T$  operators which are later used in solving the amplitudes for the  $S_v$  operators following the expressions

$$\langle \Phi_0^K | \{\widehat{H}e^T\} | \Phi_0 \rangle = \delta_{0,K} \Delta E_{\text{corr}} \quad (14)$$

and

$$\begin{aligned}\langle \Phi_v^K | \{\widehat{H}e^T\} \{1 + S_v\} | \Phi_v \rangle &= \langle \Phi_v^K | 1 + S_v | \Phi_v \rangle \langle \Phi_v | \{\widehat{H}e^T\} \{1 + S_v\} | \Phi_v \rangle \\ &= \langle \Phi_v^K | \delta_{v,K} + S_v | \Phi_v \rangle \Delta E_v,\end{aligned}\quad (15)$$

where  $K = 1, 2 \dots$  represent the singly, doubly etc. excited configurations with respect to their corresponding reference states, respectively,  $\widehat{H}e^T$  denotes the connected terms of the Dirac–Coulomb (DC) Hamiltonian ( $H$ ) with the  $T$  operators,  $\Delta E_{\text{corr}}$  and  $\Delta E_v$  are the correlation energy for the closed-core and negative of the ionization potential (IP) of the valence electron  $v$ , respectively. We consider only the singly and doubly excited configurations by defining  $T$  and  $S_v$  operators as

$$T = T_1 + T_2 \quad \text{and} \quad S_v = S_{1v} + S_{2v}. \quad (16)$$

However, we construct triple excitation configurations perturbatively by taking contractions between  $T_2$  and  $S_{2v}$  operators with  $H$  and include their contributions in the  $\Delta E_v$  evaluation. This procedure is commonly known as the CCSD(T) method.

Similar to the wave function calculation, we also calculate the scalar polarizabilities as a sum of three contributions

$$\alpha_v^{Q_L^\lambda} = \alpha_v^{Q_L^\lambda}(c) + \alpha_v^{Q_L^\lambda}(vc) + \alpha_v^{Q_L^\lambda}(v), \quad (17)$$

where  $\alpha_v^{Q_L^\lambda}(c)$ ,  $\alpha_v^{Q_L^\lambda}(vc)$  and  $\alpha_v^{Q_L^\lambda}(v)$  are referred to as the core, core-valence and valence correlation contributions, respectively.

We determine the valence correlation contributions to the polarizability in the sum-over-states approach by evaluating their matrix elements and using the experimentally known energies. As the contributions from the core and core-valence correlations to these quantities are generally small and the sum-over-states method cannot be used to determine them, we account these contributions through the following expression:

$$\alpha_v^{Q_L^\lambda} = \frac{2}{\alpha^{2(\lambda-1)}} \langle \Psi_v | Q_L^\lambda | \Psi_v^{(1)} \rangle, \quad (18)$$

which is obtained from the third-order many-body theory (MBPT(3)), where  $|\Psi_v^{(1)}\rangle$  is the first-order wave function to  $|\Psi_v^{(0)}\rangle$  due to  $Q_L^\lambda$ . The reduced matrix elements for any physical operator is evaluated using the following expression:

$$\frac{\langle \Psi_f | Q_L^\lambda | \Psi_i \rangle}{\sqrt{\langle \Psi_f | \Psi_f \rangle \langle \Psi_i | \Psi_i \rangle}} = \frac{\langle \Phi_f | \{1 + S_f^\dagger\} \overline{Q_L^\lambda} \{1 + S_i\} | \Phi_i \rangle}{\sqrt{\mathcal{N}_f \mathcal{N}_i}}, \quad (19)$$

where  $\overline{Q_L^\lambda} = e^{T^\dagger} Q_L^\lambda e^T$  and  $\mathcal{N}_v = \langle \Phi_v | e^{T^\dagger} e^T + S_v^\dagger e^{T^\dagger} e^T S_v | \Phi_v \rangle$  involve two non-truncating series in the above expression. The procedure for calculating these expressions are discussed in [8,9,11,12].

#### 4. Results and discussions

In table 1, we present the  $M1$  polarizabilities of the states associated with the clock transitions in both  $\text{Ba}^+$  and  $\text{Ra}^+$ . These results are very small in the ground states of these alkaline earth ions and mainly come from core correlations. Compared to the results for the corresponding ground and  $d_{5/2}$  states in the clock transitions of  $\text{Ca}^+$  and  $\text{Sr}^+$ , the reported results are smaller in magnitudes. These results also mainly come from their fine structures. The  $M1$  transition amplitudes between these fine-structure levels are typically of the same order of magnitudes. But the  $M1$  polarizability is larger in  $\text{Ca}^+$  and decreases gradually with respect to the size of the alkaline earth ions because fine-structure intervals increase with the size. Since the fine structure splittings are relativistic in origin, it supports the fact that the relativistic effects are large in heavy atomic systems. No other experimental or theoretical results for these quantities are available so that we can compare these with our results.

The  $E2$  polarizability for the same states in  $\text{Ba}^+$  and  $\text{Ra}^+$  are also reported in table 2. Unlike the  $M1$  polarizability results, there is no particular trend of the  $E2$  polarizabilities of the ground states with respect to the size of the systems; the polarizability increases from  $\text{Ca}^+$  to  $\text{Ba}^+$  and it comes down again for  $\text{Ra}^+$ . However, these results decrease gradually for the  $d_{5/2}$  states from  $\text{Ca}^+$  to  $\text{Ra}^+$ .

**Table 1.** The  $M1$  polarizabilities of the ground and excited states in  $Ba^+$  and  $Ra^+$  (in a.u.). The calculated amplitudes of the corresponding  $|\langle \psi_n || Q_0^1 || \psi_m \rangle|/\alpha$  matrix elements are given (in a.u.).

Transition	Amplitude	$\alpha^{M1}$	Transition	Amplitude	$\alpha^{M1}$
$\alpha_v$			$\alpha_v$		
$6s_{1/2} \rightarrow 7s_{1/2}$	0.0009	$1.4 \times 10^{-6}$	$7s_{1/2} \rightarrow 8s_{1/2}$	0.0012	$2.4 \times 10^{-6}$
	0.0015	$2.8 \times 10^{-6}$		0.0014	$2.4 \times 10^{-6}$
	0.0025	$6.9 \times 10^{-6}$		0.0021	$4.8 \times 10^{-6}$
	0.0007	$7.4 \times 10^{-6}$		0.0030	$5.4 \times 10^{-6}$
	0.0002	$6.4 \times 10^{-8}$		0.0009	$1.2 \times 10^{-6}$
	0.0001	$1.2 \times 10^{-8}$		0.0005	$3.0 \times 10^{-7}$
	$\sim 0.0$	$\sim 0.0$		0.0001	$1.1 \times 10^{-8}$
$\alpha_c$		$3.5 \times 10^{-4}$	$\alpha_c$		$6.3 \times 10^{-3}$
$\alpha_{cv}$		$\sim 0.0$	$\alpha_{cv}$		$\sim 0.0$
$\alpha_{tail}$		$\sim 0.0$	$\alpha_{tail}$		$\sim 0.0$
$\alpha_{total}$	$3.6 \times 10^{-4}$		$\alpha_{total}$		$6.4 \times 10^{-3}$
$\alpha_v$			$\alpha_v$		
$5d_{5/2} \rightarrow 5d_{3/2}$	1.5445	-72.63	$6d_{5/2} \rightarrow 6d_{3/2}$	1.5480	-35.23
	0.0194	$2.3 \times 10^{-4}$		0.0445	$1.4 \times 10^{-3}$
	0.0108	$5.3 \times 10^{-5}$		0.0236	$2.8 \times 10^{-4}$
	0.0096	$3.7 \times 10^{-5}$		0.0218	$2.1 \times 10^{-4}$
	0.0003	$5.4 \times 10^{-8}$		0.0028	$5.4 \times 10^{-6}$
	0.0024	$2.6 \times 10^{-6}$		0.0044	$9.8 \times 10^{-6}$
	0.0034	$4.6 \times 10^{-6}$		0.0035	$5.5 \times 10^{-6}$
$\alpha_c$		$3.5 \times 10^{-4}$	$\alpha_c$		$6.3 \times 10^{-3}$
$\alpha_{cv}$		$-1.4 \times 10^{-5}$	$\alpha_{cv}$		$-2.7 \times 10^{-5}$
$\alpha_{tail}$		$\sim 0.0$	$\alpha_{tail}$		$\sim 0.0$
$\alpha_{total}$	-71.63		$\alpha_{total}$		-35.23

The largest contribution to the ground-state  $E2$  polarizabilities comes from the  $d_{5/2}$  states followed by their fine-structure levels. Contributions from the core and higher continuum states (given as tail contributions) are non-negligible, but their contributions are estimated only at the MBPT(3) approximation; a better method may modify these contributions. The highest contributions to the  $d_{5/2}$  states come from their corresponding fine-structure levels followed by the ground states. These contributions are of opposite signs with respect to the contributions from the higher states and core correlation contributions for which the final values are the results of the net cancellations. Due to this reason, the signs of the ground-state polarizabilities and  $d_{5/2}$  state polarizabilities differ; this enhances the values of the differential polarizabilities between these two states.

Three experimental results available for the ground-state  $E2$  polarizability in  $Ba^+$  are: 4420(250) [17], 2462(361) [18] and 2050(100) [19] in a.u. against our result 4233.16 a.u. which is within the error bar of the first one. In fact, the  $E2$  matrix elements of  $6s \rightarrow 5d$  in  $Ba^+$  are well studied and used to estimate the lifetimes of the  $5d$  states

**Table 2.** The  $E2$  polarizabilities of the ground and excited states in  $Ba^+$  and  $Ra^+$  (in a.u.). The calculated amplitudes of the corresponding  $|\langle \psi_n || Q_1^2 || \psi_m \rangle|$  matrix elements are given (in a.u.).

Transition	Amplitude	$\alpha^{E2}$	Transition	Amplitude	$\alpha^{E2}$	
$\alpha_v$			$\alpha_v$			
$6s_{1/2} \rightarrow$	$5d_{3/2}$	12.759	$7s_{1/2} \rightarrow$	$6d_{3/2}$	14.893	805.66
	$6d_{3/2}$	16.585		$7d_{3/2}$	13.942	175.04
	$7d_{3/2}$	5.727		$8d_{3/2}$	5.631	22.55
	$8d_{3/2}$	4.036		$9d_{3/2}$	4.510	13.08
	$5d_{5/2}$	15.991		$6d_{5/2}$	19.071	1161.65
	$6d_{5/2}$	19.988		$7d_{5/2}$	16.137	232.13
	$7d_{5/2}$	7.024		$8d_{5/2}$	6.762	32.39
	$8d_{5/2}$	5.022		$9d_{5/2}$	5.615	20.23
$\alpha_c$		48.51	$\alpha_c$		64.37	
$\alpha_{cv}$		-0.15	$\alpha_{cv}$		-0.11	
$\alpha_{tail}$		10.45	$\alpha_{tail}$		16.22	
$\alpha_{total}$		4233.16	$\alpha_{total}$		2543.22	
$\alpha_v$			$\alpha_v$			
$5d_{5/2} \rightarrow$	$6s_{1/2}$	15.991	$6d_{5/2} \rightarrow$	$7s_{1/2}$	19.071	-645.36
	$7s_{1/2}$	6.409		$8s_{1/2}$	11.015	99.75
	$8s_{1/2}$	1.992		$9s_{1/2}$	2.838	4.32
	$9s_{1/2}$	1.723		$10s_{1/2}$	2.318	2.47
	$5d_{3/2}$	6.832		$6d_{3/2}$	8.786	-1134.87
	$6d_{3/2}$	5.754		$7d_{3/2}$	7.363	37.77
	$7d_{3/2}$	2.030		$8d_{3/2}$	2.452	3.06
	$8d_{3/2}$	1.457		$9d_{3/2}$	1.840	1.51
	$6d_{5/2}$	11.264		$7d_{5/2}$	13.956	133.80
	$7d_{5/2}$	4.029		$8d_{5/2}$	4.832	11.81
	$8d_{5/2}$	2.925		$9d_{5/2}$	3.744	6.25
	$5g_{7/2}$	2.965		$5g_{5/2}$	4.490	9.75
	$6g_{7/2}$	3.006		$6g_{5/2}$	4.284	8.02
	$5g_{9/2}$	10.484		$5g_{7/2}$	15.875	121.92
	$6g_{9/2}$	10.629		$6g_{7/2}$	15.148	100.25
$\alpha_c$		48.51	$\alpha_c$		64.37	
$\alpha_{cv}$		-0.87	$\alpha_{cv}$		-0.58	
$\alpha_{tail}$		12.06	$\alpha_{tail}$		18.91	
$\alpha_{total}$		-1309.39	$\alpha_{total}$		-1156.85	

precisely, implying that these amplitudes are highly accurate [20,21]. These two states contribute about 3444.09 a.u. to the ground-state  $E2$  polarizability against the reported result 1828(88) a.u. in [17]. This suggests that the results reported in [18] and [19] do not seem to be reasonable. No experimental data of the  $5d$  states are available to compare with our calculated results. Similarly, no experimental results are available for both

the ground- and excited-state  $E2$  polarizabilities in  $\text{Ra}^+$  to compare with our calculated results. In an earlier work, we had reported the ground-state  $E2$  polarizability of  $\text{Ra}^+$  as 2547.52 a.u. by considering contributions only from a few excited states in the sum of eq. (5) against the present result which is 2543.22 a.u. Both the results are in good agreement and hence, we believe that our results for the  $d_{5/2}$  states should also be accurate.

After substituting the above results, we obtain the contribution to the BBR shifts to the  $6s \rightarrow 5d_{5/2}$  transition in  $\text{Ba}^+$  through the  $M1$  and  $E2$  multipoles are  $3.3 \times 10^{-5}$  and  $3.6 \times 10^{-7}$  Hz, respectively. Similarly, contributions to the BBR shifts from the  $M1$  and  $E2$  multipoles to the  $7s \rightarrow 6d_{5/2}$  transition in  $\text{Ra}^+$  are  $1.6 \times 10^{-5}$  and  $2.4 \times 10^{-7}$  Hz, respectively. Similar to our earlier studies, the  $M1$  contributions dominate over the  $E2$  contributions. The contributions from the  $E1$  multipoles are about 0.64 Hz in  $\text{Ba}^+$  and 0.19 Hz in  $\text{Ra}^+$  for the corresponding transitions, respectively [11]. Therefore, achieving the  $10^{-18}$  precision uncertainty for the proposed atomic clock frequencies in  $\text{Ba}^+$  and  $\text{Ra}^+$  seems to be feasible as the contributions from the higher multipoles to the net BBR shifts are found to be comparatively smaller in size.

## 5. Conclusion

Following the proposals of achieving  $10^{-18}$  precision uncertainty in the  $6s \rightarrow 5d_{5/2}$  and  $7s \rightarrow 6d_{5/2}$  clock transitions in  $\text{Ba}^+$  and  $\text{Ra}^+$ , respectively, we have analysed the contributions from the  $M1$  and  $E2$  multipoles to the BBR shifts in the corresponding above clock transitions. Our study reveals that the uncertainties due to the higher multipolar BBR shifts in the proposed  $\text{Ba}^+$  and  $\text{Ra}^+$  clock transitions are below  $10^{-18}$  precision level. Therefore, these ions would be two more promising candidates for new atomic clocks which need to be undertaken by the experimentalists to measure their clock frequencies.

## Acknowledgements

The author gratefully acknowledges Bindiya Arora, Dillip Nandy and B P Das for many useful discussions. The author also thanks Ashok Vudayagiri for inviting him to present a seminar on atomic clocks at TC-2012, Hyderabad. Computations were carried out using 3TFLOP HPC Cluster at the Physical Research Laboratory, Ahmedabad.

## References

- [1] W H Oskay, S A Diddams, E A Donley, T M Fortier, T P Heavner, L Hollberg, W M Itano, S R Jefferts, M J Delaney, K Kim, F Levi, T E Parker and J C Bergquist, *Phys. Rev. Lett.* **97**, 020801 (2006)
- [2] K Matsubara, K Hayasaka, Y Ling, H Ito, S Nagano, M Kajita and M Hosokawa, *Appl. Phys. Express* **1**, 067011 (2008)
- [3] P Dube, A A Madej, J E Bernard, L Marmet, J S Boulanger and S Cundy, *Phys. Rev. Lett.* **95**, 033001 (2005)
- [4] C W Chou, D B Hume, J C J Koelemeij, D J Wineland and T Rosenband, *Phys. Rev. Lett.* **104**, 070802 (2010)
- [5] T Schneider, E Peik and Chr Tamm, *Phys. Rev. Lett.* **94**, 230801 (2005)



- [6] T Rosenband *et al*, *Science* **319**, 1808 (2008)
- [7] J A Sherman, W Trimble, S Metz, W Nagourney and N Fortson, *Progress on indium and barium single ion optical frequency standards*, 2005 Digest of the LEOS Summer Topical Meetings, IEEE Cat. No. 05TH8797 (IEEE, New York, 2005)
- [8] B K Sahoo, *Phys. Rev. A* **74**, 020501(R) (2006)
- [9] B K Sahoo, B P Das, R K Chaudhuri, D Mukherjee, R G E Timmermans and K Jungmann, *Phys. Rev. A* **76**, 040504(R) (2007)
- [10] O O Versolato, L W Wansbeek, K Jungmann, R G E Timmermans, L Willmann and H W Wilschut, *Phys. Rev. A* **83**, 043829 (2011)
- [11] B K Sahoo, R G E Timmermans, B P Das and D Mukherjee, *Phys. Rev. A* **80**, 062506 (2009)
- [12] B K Sahoo, B P Das and D Mukherjee, *Phys. Rev. A* **79**, 052511 (2009)
- [13] M Kallay, H S Nataraj, B K Sahoo, B P Das and L Visscher, *Phys. Rev. A* **83**, 030503 (2011)
- [14] B Arora, D K Nandy and B K Sahoo, *Phys. Rev. A* **85**, 012506 (2012)
- [15] S H Autler and C H Townes, *Phys. Rev.* **100**, 703 (1955)
- [16] J W Farley and W H Wing, *Phys. Rev. A* **23**, 2397 (1981)
- [17] E L Snow and S R Lundeen, *Phys. Rev. A* **76**, 052505 (2007)
- [18] E L Snow, M A Gearba, R A Komara, S R Lundeen and W G Sturuss, *Phys. Rev. A* **71**, 022510 (2005)
- [19] T F Gallagher, R Kachru and N H Tran, *Phys. Rev. A* **26**, 2611 (1982)
- [20] B K Sahoo, Md R Islam, B P Das, R K Chaudhuri and D Mukherjee, *Phys. Rev. A* **74**, 062504 (2006)
- [21] B K Sahoo, B P Das, R K Chaudhuri and D Mukherjee, *Phys. Rev. A* **75**, 032507 (2007)