Blackbody-radiation shift in a $^{88}$Sr$^+$ ion optical frequency standard

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Abstract

The blackbody-radiation (BBR) shift of the 5s–4d$^5$/$^2$ clock transition in $^{88}$Sr$^+$ is calculated to be $0.250(9)$ Hz at room temperature, $T = 300$ K, using the relativistic all-order method where all single and double excitations of the Dirac–Fock wavefunction are included to all orders of perturbation theory. The BBR shift is a major component in the uncertainty budget of the optical frequency standard based on the $^{88}$Sr$^+$ trapped ion. The scalar polarizabilities of the 5s and 4d$^5$/$^2$ levels, as well as the tensor polarizability of the 4d$^5$/$^2$ level, are presented together with the evaluation of their uncertainties. The lifetimes of the 4d$^3$/$^2$, 4d$^5$/$^2$, 5p$^1$/$^2$ and 5p$^3$/$^2$ states are calculated and compared with experimental values.

1. Introduction

The current definition of a second in the International System of Units (SI) is based on the microwave transition between the two hyperfine levels of the ground state of $^{133}$Cs. The present relative standard uncertainty of Cs microwave frequency standard is around $4 \times 10^{-16}$ [1]. Since the frequencies of feasible optical clock transitions are five orders of magnitude greater than the standard microwave transitions, optical frequency standards may achieve even smaller relative uncertainties. Significant recent progress in optical spectroscopy and measurement techniques has led to the achievement of relative standard uncertainties in optical frequency standards that are comparable to the Cs microwave benchmark.

In 2006, the International Committee for Weights and Measures (CIPM) recommended that the following transitions frequencies shall be used as secondary representations of the second [2]: ground-state hyperfine microwave transition in $^{87}$Rb [3, 4], 5s$^2$S$_{1/2}$–4d$^2$D$_{5/2}$ optical transition of the $^{88}$Sr$^+$ ion [5, 6], 5d$^{10}$6s$^2$S$_{1/2}$ (F = 0)–5d$^4$6s$^2$2D$_{3/2}$ (F = 2) optical transition in the $^{199}$Hg$^+$ ion [7, 8], 6s$^2$S$_{1/2}$ (F = 0)–5d$^4$D$_{3/2}$ (F = 2) optical transition in the $^{171}$Yb$^+$ ion [9, 10] and 5s$^2$1S$_0$–5s5p $^3$P$^0$ transition in the $^{87}$Sr neutral atom [11–13]. With extremely low systematic perturbations and better stability and accuracy, such optical frequency standards can reach a systematic fractional uncertainty of the order of $10^{-18}$ [14, 6]. More precise frequency standards will open ways to improve global positioning systems and tracking of deep-space probes, and perform more accurate measurements of the fundamental constants and testing of physics postulates.

In this paper, we treat one of the optical transitions recommended as the secondary standard: the 5s$^2$S$_{1/2}$–4d$^2$D$_{5/2}$ electric-quadrupole transition of $^{88}$Sr$^+$ at 445 THz (674 nm). The reported frequency measurements of this transition in a single trapped $^{88}$Sr$^+$ ion have achieved a spectral resolution of better than 1.5 Hz [5, 6, 15, 16]. Methods based on this transition have the potential to reduce relative systematic uncertainty to the level of $10^{-17}$ or below [6].

The accuracy of optical frequency standards is limited by the frequency shift in the clock transition caused by the interactions of the ion with external fields. The linear Zeeman shift in the $^{88}$Sr$^+$ system can be eliminated by use of the line centre of symmetric Zeeman states; the second-order Zeeman shift is around 1 mHz and is negligible at the current level of precision [5]. The second-order Doppler shifts due to micromotion of the trapped ion are estimated to be less than 0.01 Hz [6]. The major contributions to the systematic frequency shifts come from Stark shifts with the blackbody radiation (BBR) Stark shift being one of the most important contributions at room temperature. Experimental measurements of the BBR radiation shifts are difficult. Here, we present theoretical calculations that result in an estimate
of this shift that reduces the previous uncertainty [5, 6] by a factor of 10.

In this paper, we present a relativistic all-order calculation of the static polarizabilities of the 5s1/2 and 4d5/2 states of 88Sr. The relativistic all-order method used here is one of the most accurate methods used for the calculation of atomic properties of monovalent systems. Readers are referred to [17] and references therein for a review of this method and its applications. We use these polarizability values to evaluate the BBR shift of the clock transition at room temperature.

The dynamic correction to the electric-dipole contribution and multipolar corrections due to M1 and E2 transitions are incorporated. Uncertainty of the final BBR shift is estimated to be 3.6%. Lifetimes of the low-lying excited 4d3/2, 4d5/2, 5p1/2 and 5p3/2 states are also calculated and compared with experiments.

2. Method

The electrical field $E$ radiated by a blackbody at temperature $T$, as given by Planck’s law,

$$E^2(\omega) d\omega = \frac{8a^3}{\pi} \frac{\omega^3 d\omega}{\exp(\omega/k_B T) - 1},$$

induces a nonresonant perturbation of the optical transition at room temperature [18]. Assuming that the system evolves adiabatically, the frequency shift of an atomic state due to such an electrical field can be related to the static electric-dipole polarizability $\alpha_0$ by (see [19])

$$\Delta_0 = -\frac{1}{2} \left( 831.9 \text{ V m}^{-1} \right)^2 \left( \frac{T(K)}{300} \right)^4 a_0(1 + \eta).$$

Here $\eta$ is a small dynamic correction due to the frequency distribution. Only the electric-dipole transition part of the contribution is considered in the formula because the contributions from M1 and E2 transitions are suppressed by a factor of $\alpha^2$ [19]. We estimate these multipolar corrections together with the dynamic correction $\eta$ in section 5 of this work. The overall BBR shift of the clock transition frequency is then calculated as the difference between the BBR shifts of the individual levels involved in the transition:

$$\Delta_{BBR} (5s \rightarrow 4d_{5/2}) = -\frac{1}{2} [a_0(4d_{5/2}) - a_0(5s_{1/2})]$$

$$\times (831.9 \text{ V m}^{-1})^2 \left( \frac{T(K)}{300} \right)^4.$$

Therefore, the evaluation of the BBR shift requires accurate calculation of static scalar polarizabilities of 88Sr in the 5s1/2 ground and 4d5/2 excited states. The static tensor polarizability of the 4d5/2 state is also calculated in the present work though the effect of the tensor part of polarizability is averaged out due to the isotropic nature of the electric field radiated by the blackbody.

The calculation of the scalar polarizability of a monovalent atom can be separated into three parts: the contribution of the electrons in the ionic core, $\alpha_{\text{core}}$; a small term, $\alpha_{\text{vc}}$, that changes the core polarizability due to the presence of the valence electron and the dominant contribution, $\alpha_{\text{vc}}$, from the valence electron. The ionic core polarizability used here was calculated using the random-phase approximation (RPA) [20]. We calculate the $\alpha_{\text{vc}}$ contribution in the RPA as well for consistency with the ionic core value. The valence scalar $\alpha_0$ and tensor $\alpha_2$ polarizabilities of an atom in a state $\nu$ can be expressed as the sum over all intermediate states $k$ allowed by the electric-dipole selection rules:

$$\alpha_0 = \frac{2}{3(2j_k + 1)} \sum_k \langle k || D || \nu \rangle^2 E_k - E_\nu,$$

$$\alpha_2 = -4C \sum_k (-1)^j_k j_k C_k \left( \frac{k || D || \nu}{E_k - E_\nu} \right)^2,$$

where $\langle k || D || \nu \rangle$ are the reduced electric-dipole (E1) matrix elements, and $E_j$ is the energy of the $j$th state. We also separate the valence polarizability into two parts, the main term $\alpha_{\text{main}}$ containing the first few dominant contributions and the remainder $\alpha_{\text{tail}}$. We use electric-dipole matrix elements calculated using the relativistic single–double (SD) all-order method (see [17] for detail) and experimental energies from [21] for the calculations of the main term. Triple excitations are included partially where needed; the resulting values are referred to as SDptT (single, double, partial triple) data.

The tail contribution for the 5s state is negligible and is estimated in the lowest-order Dirac–Fock (DF) approximation. Significantly, larger tail contribution to the 4d5/2 polarizability is evaluated in both the DF and the random phase (RP) approximations and scaled to account for other missing correlation corrections.

In this work, we use atomic units (au), in which, $e$, $m_e$, $4\pi \epsilon_0$ and the reduced Planck constant $\hbar$ have the numerical value 1. Polarizability in au has the dimension of volume, and its numerical values presented here are thus expressed in units of $a_0^3$, where $a_0 \approx 0.052918$ nm is the Bohr radius. The atomic units for $\alpha$ can be converted to SI units via $\alpha/\hbar$ (Hz (Vm$^{-1}$)$^{-2}$) $= 2.48832 \times 10^{-8} \times \epsilon_0 a_0^3 / \hbar$, and the Planck constant $\hbar$ is factored out.

We have used the B-spline method to construct a finite basis set for radial Dirac equations as introduced in [22]. Seventy B-splines of order $k = 8$ are constrained to a spherical cavity with $R = 220$ au for each angular momentum. Such a large cavity is chosen to accurately evaluate as many 4d5/2–n5f1/2 transitions as practically possible to reduce the uncertainty in the remainder.

3. Polarizabilities

Table 1 shows the contributions of the individual transitions to the ground-state scalar polarizability $\alpha_0$. The main contributions are listed separately along with the respective values of the electric-dipole matrix elements. The tail contributions are grouped together as $\alpha_{\text{tail}}$. For the main contributions, we use our ab initio SD all-order values of the matrix elements and experimental energies from [21]. The 5s1/2–5p1/2 and 5s1/2–5p3/2 transitions contribute over 99.9%
to the valence polarizability and 94% of the total polarizability value. The same calculation of these transitions in Rb agrees with the high-precision experiment to 0.26% [23]. In fact, the SD values for the primary $ns$–$np$ transitions in Li, Na, K, Rb and Cs agree with various types of high-precision experiments to 0.1%–0.4% [23]. There is no reason to expect reduced accuracy in the case of Sr*, and we take the uncertainty of these matrix element values to be 0.5%. Unfortunately, we know of no way to accurately estimate the missing additional contributions to the dominant correlation correction to these transitions, unlike the case of the $4d$–$5p$ and $5s$–$4d$ transitions, where semi-empirical scaling makes possible an uncertainty estimate that does not directly depend upon comparison with the experiment.

The core contribution taken from [20] is estimated to be accurate to 5%, based on the comparison of the RPA and experimental polarizability values for noble gases. The tail contribution is calculated using the DF approximation and is negligible in comparison with the total polarizability. The error of the tail is taken to be 100%. As a result, all uncertainties except those associated with the $5s$–$5p$ matrix elements are negligible. The resulting final uncertainty of the 5s polarizability is thus estimated to be 1%. We note that accurate measurement of either $5s$–$5p$ oscillator strengths or 5p lifetimes ($5p$–$4d$ contributions are small and can be accurately calculated) will help to significantly reduce this uncertainty.

Table 2 shows the contributions from the individual transitions to the $4d_{5/2}$ polarizability. Three types of transitions contribute to the $4d_{5/2}$ polarizability: $4d_{5/2}$–$np_{3/2}$, $4d_{5/2}$–$nf_{3/2}$ and $4d_{5/2}$–$nf_{7/2}$. The sum over the $4d_{5/2}$–$np_{3/2}$ transitions converges very quickly with the $4d_{5/2}$–$5p_{3/2}$ term being overwhelmingly dominant. We obtain an accurate value for this matrix element using a semi-empirical scaling procedure that evaluates some classes of correlation corrections omitted by the current all-order calculations. The scaling procedure is described in [24, 25]. Briefly, the single valence excitation coefficients are multiplied by the ratio of the corresponding experimental and theoretical correlation energies, and the matrix element calculation is repeated with the modified excitation coefficients. The scaling procedure is particularly suitable for this transition because the matrix element contribution containing the single valence excitation coefficients is dominant in this case (but not for the $5s$–$5p$ matrix elements discussed earlier). We conduct the scaling starting from both the SD and SDpT approximations. The scaling factors for the SD and SDpT calculations are different, and we take the scaled SD value as the final result for the $4d_{5/2}$–$5p_{3/2}$ matrix element, based on the comparisons of similar calculations in alkali-metal atoms with experiments [26–29]. The absolute values of the reduced $4d_{5/2}$–$5p_{3/2}$ matrix elements calculated in different approximations are summarized in table 3, together with four other transitions that represent similar cases. The uncertainties are determined as the maximum difference between the scaled SD values and the ab initio SDpT and scaled SDpT values. A notable feature of this table is a close agreement of the scaled SD and SDpT results. The sum of the contributions from the $4d_{5/2}$–$nf_{3/2}$ and $4d_{5/2}$–$nf_{7/2}$ transitions converges slowly; therefore, we include

| Transition | DF | SD | SDpT | $\alpha_{sc}$ | $\alpha_{SDpTsc}$ | Final |
|-----------|----|----|------|----------------|------------------|-------|
| $4d_{5/2}$–$5p_{3/2}$ | 5.002 | 4.150 | 4.198 | 4.187 | 4.173 | 4.187(14) |
| $4d_{5/2}$–$4f_{5/2}$ | 0.964 | 0.779 | 0.790 | 0.789 | 0.788 | 0.789(4) |
| $4d_{5/2}$–$4f_{7/2}$ | 4.313 | 3.486 | 3.536 | 3.528 | 3.509 | 3.528(19) |
| $4d_{5/2}$–$nf_{3/2}$ | 3.729 | 3.083 | 3.119 | 3.112 | 3.102 | 3.112(10) |
| $4d_{5/2}$–$nf_{7/2}$ | 1.657 | 1.369 | 1.386 | 1.383 | 1.378 | 1.383(5) |

Table 3. Reduced electric-dipole transition matrix elements calculated using different approximations: the Dirac–Fock (DF), single–double all-order method (SD) and the single–double all-order method including partial triple-excitation contributions (SDpT); the label ‘sc’ indicates the corresponding scaled values. All values are given in atomic units.
as many transitions as realistically possible in the main term calculation. Scaled values are used for the 4d5/2–4f5/2 and 4d5/2–4f7/2 transitions in the polarizability calculations.

The tail contribution of the 4d5/2–nf7/2 terms is particularly large; its DF value (3.5 au) is 5% of the total polarizability. Therefore, we carry out several additional calculations to accurately evaluate the tail contribution and estimate its uncertainty. Since the largest part of the correlation correction for the 4d5/2–nf7/2 transitions with n > 9 terms comes from RPA-like terms, the RPA is expected to produce a better result than the DF one. We carried out the RPA calculation of the tail and obtained a lower value of 2.9 au. We also calculated the last few terms using the DF and RP approximations and compared the results with our all-order values. We found that the DF and RP approximations overestimate the polarizability contributions by 35% and 28%, relative to the DF and RPA values. To improve our accuracy, we scale both the DF and RPA results by these respective amounts to obtain a DF-scaled value of 2.26 au and a RPA-scaled value of 2.06 au. We take the RPA-scaled value as the final one and the difference of these two values as its uncertainty.

We also list the contributions from various transitions to the 4d5/2 tensor polarizability α2 in table 2. The 4d5/2–np3/2 transition gives the dominant contribution to the tensor polarizability. The tail contribution is smaller yet significant and is obtained by the same procedure as the tail of the scalar 4d5/2 polarizability.

4. Lifetimes

The contributions to the lifetimes of the 5p1/2 and 5p3/2 states are given in table 4. Experimental energies from [21] are used in the evaluation of the transition rates. The lifetime is calculated as the inverse of the sum of the appropriate Einstein A-coefficients, which are proportional to the square of the dipole matrix elements, experimental energies from [21] are used. Our results are in excellent agreement with experimental lifetimes τ(5p1/2) = 7.39(7) ns and τ(5p3/2) = 6.63(7) ns by Pinnington et al measured using a laser-induced fluorescence method [30], and τ(5p1/2) = 7.47(7) ns and τ(5p3/2) = 6.69(7) ns by Kuske et al measured using the fast-beam-laser technique [31].

As a further test of accuracy of our approach and accuracy of our all-order 4d wavefunctions, we carry out the calculation of the 4d3/2 and 4d5/2 lifetimes that requires evaluation of the electric-quadrupole and magnetic-dipole transitions.

The lifetime of the 4d3/2 state is calculated to be 0.441(3) s, where the main contribution comes from the E2(4d3/2–5s1/2) = 11.13(3) au matrix element. The contribution from 4d3/2–5s1/2 M1 transition is evaluated to be negligible. The most recent lifetime measurements of the 4d3/2 states of Sr+ include 0.435(4) s result obtained by using optical pumping and the 0.455(29) s value obtained by using laser probing as reported in the same work [32]. Our result agrees with the experimental results within the uncertainty limits. The lifetime of the 4d5/2 state is calculated to be 0.394(3) s. The contribution to the A-coefficients from the 4d5/2–5s1/2 E2 transition overwhelmingly dominates, and the corresponding reduced matrix element is 13.75(4) au. The reduced matrix element for the 4d5/2–4d3/2 E2 transition is 5.98(2) au, but its contribution to the lifetime is negligible due to the small energy interval between these two states. Our value for the 4d5/2–4d3/2 M1 reduced matrix element, 1.55 au, is in agreement with the result from [34]. The contributions from M1 transitions only affect the third decimal of the lifetime result of the 4d5/2 state, and can be neglected at the present level of accuracy. The two-photon transitions contribute 0.03% to the 4d lifetimes. Our result is found in good agreement with the lifetime measurements of 0.3908(16) s and 0.408(22) s done by Letchumanan et al [37] and Biemont et al [32], respectively. More experimental and theoretical results for the lifetimes of these two states are given in table 5.

5. BBR shift

We use our scalar polarization values to evaluate the shift in the clock transition in 88Sr+ due to blackbody radiation at T = 300 K to be 0.252(9) Hz. The dynamic correction [19] is estimated to be η = 0.0013 and η = 0.0064 for the 5s and 4d3/2 states, respectively. The resulting correction to the BBR shift is −0.002 Hz and our final value for the BBR shift is 0.250(9) Hz. The overall uncertainty in the final result comes from the uncertainty in the values of the 5s–5p3/2 and 5p3/2–4d3/2 matrix elements, and 4d5/2–nf7/2 tail. The first two sources of the uncertainties may be removed if these values were determined experimentally. The 5s–5p3/2 and 5p3/2–4d3/2 matrix elements can be obtained via either lifetime, ground-state polarizability, oscillator strength or light shift ratio measurements [41], with the first two types of experiments useful for the first matrix element and the second

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Table 4. Contributions to the lifetimes of the 5p1/2 and 5p3/2 states. The transition rates A are given in 10^6 s^-1 and the lifetimes τ are given in ns.

| 5p1/2         | 5p3/2         |
|---------------|---------------|
| A(5p1/2–5s)   | 128.04        |
| A(5p1/2–4d3/2)| 7.54          |
| ∑A            | 135.58        |
| τ(5p1/2)      | 7.376         |
| Expt [30]     | 7.39(7)       |
| Expt [31]     | 7.47(7)       |

Table 5. Lifetimes of the 4d3/2 and 4d5/2 levels (s).

| Levels       | Experiment | Other calculations | Present |
|--------------|------------|--------------------|---------|
| 4d3/2        | 0.435(4)   | 0.443              | 0.441(3) |
|              | 0.455(29)  | 0.426              | 0.441    |
|              | 0.455(4)   | 0.422              | 0.394(3) |
| 4d5/2        | 0.3908(16) | 0.404              | 0.396(3) |
|              | 0.408(22)  | 0.357              | 0.384    |
|              | 0.372(25)  | 0.384              | 0.396    |

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two types for both matrix elements. We note that ionic core uncertainty is not included in the BBR uncertainty since the core contribution is the same for both levels and subtracts out. The small term $\alpha_c$ that changes the core polarizability due to the presence of the valence electron is different for the 5s and 4d$^5$2 states and contributes 0.5% to the BBR shift. We also estimated the M1 and E2 contributions to the BBR shift using the approach described in [19] and found them to be negligible (below 0.01%).

In table 6, we compare our polarizability and BBR shift results with other theoretical calculations [5, 6, 33, 39, 40]. We note that our calculation is the most complete one at present. The calculation of Mitroy et al [33] was carried out by diagonalizing a semi-empirical Hamiltonian in a large-dimension single-electron basis that gives a BBR shift of 0.242 Hz. The use of experimental energies changes this result to 0.233 Hz [33]. The accuracy of the scalar polarizability of the ground state $\alpha_0 = 99.88$ au was estimated to be 2–3% in [33]. The results of Barklem and O’Mara [39] are derived from experimental oscillator strengths (note that the core polarizability of 5.8$a_0^3$ was added to the values listed in [39]). The results of Madej et al [5] are obtained mainly by summing over the transition rates calculated in [42] using the multiconfiguration Hartree–Fock (MCHF) method. Addition of the core contribution (5.8 au) to the results of [5] leads to $\alpha_0(5s) = 90.4$ au and $\alpha_0(4d^52) = 54$ au values that are in agreement with our results within the uncertainties quoted in [5]; the $\alpha_0(4d^52)$ value is lower than the present owing to the omission of the higher-order transition contributions. The scalar polarizability of the 5s$^11/2$ state calculated by Patil and Tang [40] is obtained by evaluating the transition matrix elements with simple wavefunctions based on the asymptotic behaviour and on the binding energies of the valence electron. Our result is in good agreement with their calculation. Another estimation of the BBR shift is given by Margolis et al in [6], but the approach is not stated.

6. Conclusion

In summary, we calculated the polarizabilities of the 5s and 4d$^5$2 states in $^{88}$Sr$^+$, and the value of BBR shift of the corresponding clock transition at room temperature. The dynamic correction to the electric-dipole contribution and the multipolar corrections due to M1 and E2 transitions were estimated and found to be small at the present level of accuracy. Lifetimes of the low-lying excited 4d$^5$2, 4d$^5$2, 5p$^1/2$ and 5p$^3/2$ states were also calculated and compared with experiments for further tests of our approach. The uncertainty of the final BBR value was estimated. The main contributions to the uncertainties were analysed and possible experiments were suggested to further reduce the uncertainties of the BBR shift.

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Table 6. Comparison of static scalar polarizabilities for the 5s$1/2$ and 4d$5/2$ states and BBR shift for the 5s$1/2$–4d$5/2$ transition in the $^{88}$Sr$^+$ ion at $T = 300$ K. The polarizability values are in $a_0^3$ and the BBR shift is in Hz.

|                | Present work | [33] | [39] | [5] | [40] | [6] |
|----------------|--------------|------|------|-----|-----|-----|
| $\alpha_0(5s)$ | 91.3(9)      | 93.3 | 84.6(3.6) | 91.47 |
| $\alpha_0(4d^52)$ | 62.0(5)     | 61.77 | 62.92$^a$ | 57.0 | 48(12) |
| BBR shift     | 0.250(9)    | 0.242 | 0.233$^a$ | 0.31 | 0.33(0.12) | 0.33(9) |

$^a$ Results are obtained using experimental energies.
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