

## Frequency Shift of the Cesium Clock Transition due to Blackbody Radiation

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We perform *ab initio* calculations of the frequency shift induced by a static electric field on the cesium clock hyperfine transition. The calculations are used to find the frequency shifts due to blackbody radiation. Our result [ $\delta\nu/E^2 = -2.26(2) \times 10^{-10}$  Hz/(V/m)<sup>2</sup>] is in good agreement with early measurements and *ab initio* calculations performed in other groups. We present arguments against recent claims that the actual value of the effect might be smaller. The difference ( $\sim 10\%$ ) between *ab initio* and semiempirical calculations is due to the contribution of the continuum spectrum in the sum over intermediate states.

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Atomic clocks are now important for both practical applications and fundamental physics. One of the dominant uncertainties in high-precision measurements of frequencies in atomic clocks is the ac Stark shift induced by blackbody radiation (see e.g. [1]). There is some disagreement on the value of this shift for cesium. Early measurements [2–4] and *ab initio* calculations [5,6] support a value which is close to  $-2.2 \times 10^{-10}$  Hz/(V/m)<sup>2</sup>, while more recent measurements [7,8] and semiempirical calculations [9–11] claim that the actual number might be about 10% smaller.

In the present work, we have performed fully *ab initio* calculations of the radiation frequency shift and have identified the source of the disagreement between different theoretical results as the contribution of the continuum spectrum states into the summation over the complete set of intermediate states. The continuum spectrum was included in all the *ab initio* calculations and missed in the semiempirical considerations. We demonstrate that adding the contribution of the continuum spectrum to where it was missed brings all theoretical results to good agreement with each other and with early measurements. We have performed calculations of the radiation frequency shift for other atoms and ions in Ref. [12].

Blackbody radiation creates a temperature-dependent electric field, described by the Planck radiation law

$$E^2(\omega) = \frac{8\alpha}{\pi} \frac{\omega^3 d\omega}{\exp(\omega/kT) - 1}. \quad (1)$$

This leads to the following expression for the average electric field radiated by a blackbody at temperature  $T$ :

$$\langle E^2 \rangle = (831.9 \text{ V/m})^2 [T(\text{K})/300]^4. \quad (2)$$

This electric field causes a temperature-dependent frequency shift of the atomic microwave clock transitions. It can be presented in the form (see, e.g. [1])

$$\delta\nu/\nu_0 = \beta(T/T_0)^4 [1 + \epsilon(T/T_0)^2]. \quad (3)$$

Here  $T_0$  is usually assumed to be room temperature ( $T_0 =$

300 K). The frequency shift in a static electric field is

$$\delta\nu = kE^2. \quad (4)$$

Coefficients  $k$  and  $\beta$  are related by

$$\begin{aligned} \beta &= \frac{k}{\nu_0} (831.9 \text{ V/m})^2 \\ &= k \times 7.529 \times 10^{-5} (\text{V/m})^2 \text{ Hz}^{-1} \quad (\text{for Cs}), \end{aligned} \quad (5)$$

while  $\epsilon$  is a small correction due to frequency distribution (1). In the present work, we calculate the coefficient  $k$ .

In the case when there is no other external electric field, the radiation shift can be expressed in terms of the scalar hyperfine polarizability of the atom. This corresponds to averaging over all possible directions of the electric field. The hyperfine polarizability is the difference in the atomic polarizabilities between different hyperfine structure states of the atom. The lowest-order effect is linear in the hyperfine interaction and quadratic in the electric field. The corresponding third-order perturbation theory expressions, after angular reduction, have the form

$$\begin{aligned} \delta\nu_1(as) &= e^2 \langle E^2 \rangle \frac{2I+1}{6} \\ &\times \sum_{n,m,j} \frac{A_{as,ns} \langle ns || r || mp_j \rangle \langle mp_j || r || as \rangle}{(\epsilon_{as} - \epsilon_{ns})(\epsilon_{as} - \epsilon_{mp_j})}, \end{aligned} \quad (6)$$

$$\begin{aligned} \delta\nu_2(as) &= \frac{e^2 \langle E^2 \rangle}{6} \sum_j (C_{I+1/2} - C_{I-1/2}) \\ &\times \sum_{n,m} \frac{\langle as || r || np_j \rangle A_{np_j,mp_j} \langle mp_j || r || as \rangle}{(\epsilon_{as} - \epsilon_{np_j})(\epsilon_{as} - \epsilon_{mp_j})}, \end{aligned} \quad (7)$$

and

$$\delta\nu_{\text{norm}}(as) = -e^2 \langle E^2 \rangle \frac{2I+1}{12} A_{as} \sum_{m,j} \frac{|\langle as || r || mp_j \rangle|^2}{(\epsilon_{as} - \epsilon_{mp_j})^2}. \quad (8)$$

Here

$$C_F = \sum_{F'} (2F' + 1) [F'(F' + 1) - I(I + 1) - j(j + 1)] \times \begin{Bmatrix} 1/2 & F & I \\ F' & j & 1 \end{Bmatrix}^2, \quad F' = |I - J|, I + J,$$

$A_{ns}$  is the hfs constant of the  $ns$  state,  $A_{m,n}$  is the off-diagonal hfs matrix element,  $I$  is nuclear spin,  $\mathbf{F} = \mathbf{I} + \mathbf{J}$ ,  $\mathbf{J}$  is the total electron momentum of the atom in the ground state ( $J = 1/2$ ), and  $j$  is the total momentum of virtual  $p$  states ( $j = 1/2, 3/2$ ). Summation goes over a complete set of  $ns$ ,  $mp_{1/2}$ , and  $mp_{3/2}$  states.

In order to calculate the frequency shift to the hfs transitions due to the electric field, one needs to have a complete set of states and to have the energies, electric dipole transition amplitudes, and hyperfine structure matrix elements corresponding to these states. It is possible to consider summation over the physical states and to then use experimental data to perform the calculations. The lowest valence states for which experimental data are usually available dominate in the summation. Off-diagonal hfs matrix elements can be obtained to a high accuracy as the square root of the product of corresponding hfs constants:  $A_{m,n} = \sqrt{A_m A_n}$  (see, e.g. [13]). However, the accuracy of this approach is limited by the need to include the *tail* contribution from highly excited states including states in the continuum. This contribution can be very significant, and its calculation is not easier than the calculation of the whole sum.

Therefore, in the present work, we use an *ab initio* approach in which high accuracy is achieved by the inclusion of all important many-body and relativistic effects. We make only one exception toward the semiempirical approach. The frequency shift is dominated by the term (8) which is proportional to the hfs in the ground state. It is natural to use an experimental hfs constant in the dominating term to have more accurate results. Note, however, that the difference with complete *ab initio* calculations is small.

Calculations start from the relativistic Hartree-Fock (RHF) method in the  $V^{N-1}$  approximation. This means that the initial RHF procedure is done for a closed-shell atomic core with the valence electron removed. After that, states of the external electron are calculated in the field of the frozen core. Correlations are included by means of the correlation potential method [14]. We use two different approximations for the correlation potential  $\hat{\Sigma}$ . First, we calculate it in the lowest, second-order of the many-body perturbation theory (MBPT). We use the notation  $\hat{\Sigma}^{(2)}$  for the corresponding correlation potential. Then we also include into  $\hat{\Sigma}$  two classes of the higher-order terms: screening of the Coulomb interaction and hole-particle interaction (see, e.g. [15] for details). These two effects are included in all orders of the MBPT, and the corresponding correlation potential is named  $\hat{\Sigma}^{(\infty)}$ .

To calculate  $\hat{\Sigma}^{(2)}$ , we need a complete set of single-electron orbitals. We use the  $B$ -spline technique [16,17] to construct the basis. The orbitals are built as linear

combinations of 50  $B$  splines in a cavity of radius  $40a_B$ . The coefficients are chosen from the condition that the orbitals are eigenstates of the RHF Hamiltonian  $\hat{H}_0$  of the closed-shell core. The  $\hat{\Sigma}^{(\infty)}$  operator is calculated with the technique which combines solving equations for the Green functions (for the direct diagram) with the summation over a complete set of states (exchange diagram) [15].

The correlation potential  $\hat{\Sigma}$  is then used to build a new set of single-electron states, the so-called Brueckner orbitals. This set is to be used in the summation in Eqs. (6)–(8). Here again we use the  $B$ -spline technique to build the basis. The procedure is very similar to the construction of the RHF  $B$ -spline basis. The only difference is that new orbitals are now the eigenstates of the  $\hat{H}_0 + \hat{\Sigma}$  Hamiltonian, where  $\hat{\Sigma}$  is either  $\hat{\Sigma}^{(2)}$  or  $\hat{\Sigma}^{(\infty)}$ .

Brueckner orbitals which correspond to the lowest valence states are good approximations to the real physical states. Their quality can be checked by comparing experimental and theoretical energies. Moreover, their quality can be further improved by rescaling the correlation potential  $\hat{\Sigma}$  to fit experimental energies exactly. We do this by replacing the  $\hat{H}_0 + \hat{\Sigma}$  with the  $\hat{H}_0 + \lambda \hat{\Sigma}$  Hamiltonian in which the rescaling parameter  $\lambda$  is chosen for each partial wave to fit the energy of the first valence state. The values of  $\lambda$  are  $\lambda_s = 0.8$  and  $\lambda_p = 0.85$  for  $\hat{\Sigma}^{(2)}$  and  $\lambda_s = 0.99$  and  $\lambda_p = 0.95$  for  $\hat{\Sigma}^{(\infty)}$ . Note that the values are very close to unity. This means that even without rescaling the accuracy is very good and only a small adjustment to the value of  $\hat{\Sigma}$  is needed. Note also that, since the rescaling procedure affects both energies and wave functions, it usually leads to improved values of the matrix elements of external fields. In fact, this is a semiempirical method to include omitted higher-order correlation corrections.

Matrix elements of the hfs and electric dipole operators are found by means of the time-dependent Hartree-Fock (TDHF) method [14,18]. This method is equivalent to the well-known random-phase approximation. In the TDHF method, single-electron wave functions are presented in the form  $\psi = \psi_0 + \delta\psi$ , where  $\psi_0$  is an unperturbed wave function. It is an eigenstate of the RHF Hamiltonian  $\hat{H}_0$ :  $(\hat{H}_0 - \epsilon_0)\psi_0 = 0$ .  $\delta\psi$  is the correction due to external field. It can be found by solving the TDHF equation

$$(\hat{H}_0 - \epsilon_0)\delta\psi = -\delta\epsilon\psi_0 - \hat{F}\psi_0 - \delta\hat{V}^{N-1}\psi_0, \quad (9)$$

where  $\delta\epsilon$  is the correction to the energy due to external field ( $\delta\epsilon \equiv 0$  for the electric dipole operator),  $\hat{F}$  is the operator of the external field ( $\hat{H}_{\text{hfs}}$  or  $e\mathbf{E} \cdot \mathbf{r}$ ), and  $\delta\hat{V}^{N-1}$  is the correction to the self-consistent potential of the core due to external field. The TDHF equations are solved self-

consistently for all states in the core. Then matrix elements between any (core or valence) states  $n$  and  $m$  are given by

$$\langle \psi_n | \hat{F} + \delta \hat{V}^{N-1} | \psi_m \rangle. \quad (10)$$

The best results are achieved when  $\psi_n$  and  $\psi_m$  are Brueckner orbitals calculated with rescaled correlation potential  $\hat{\Sigma}$ .

We use Eq. (10) for all hfs and electric dipole matrix elements in (6)–(8), except for the ground state hfs matrix element in (8), where we use experimental data.

To check the accuracy of the calculations, we perform calculations of the hfs in the ground state and of the static scalar polarizability. Polarizability is given by the expression

$$\alpha_0(a) = \frac{2}{3} \sum_m \frac{|\langle a || r || m \rangle|^2}{\epsilon_a - \epsilon_m}, \quad (11)$$

which is very similar to the term (8) for the frequency shift. The most important difference is that the energy denominator is squared in term (8) but not in (11). This means better convergence with respect to the summation over complete set of states for term (8) than for (11). Therefore, if good accuracy is achieved for polarizabilities, even better accuracy should be expected for the term (8) (see also Ref. [10]).

However, the behavior of the other two terms, (6) and (7), is very different and calculation of polarizabilities tells us little about accuracy for these terms. Therefore, we also perform detailed calculations of the hfs constants of the ground state. The inclusion of core polarization [second term in (10)] involves summation over the complete set of states similar to what is needed for term (6). Comparing experimental and theoretical hfs is a good test for the accuracy for this term.

In addition to term (10), we also include two smaller contributions to the hfs: structure radiation and the correction due to the change of the normalization of the wave function. Our final result for the hfs constant is 2278 MHz, which is in excellent agreement with the experimental value 2298 MHz [19]. The result for static polarizability is  $\alpha_0 = 399.0a_0^3$ , which is also in a very good agreement with the experimental value  $401.0(6)a_0^3$  [20].

Table I presents the contributions of terms (6)–(8) to the total frequency shift of the hfs transition for the ground state of  $^{133}\text{Cs}$  calculated in different approximations. Term (8) dominates, while term (7) is small but still important. Results obtained with  $\hat{\Sigma}^{(2)}$  and  $\hat{\Sigma}^{(\infty)}$  differ significantly (14%). However, after rescaling the results for both  $\hat{\Sigma}^{(2)}$  and  $\hat{\Sigma}^{(\infty)}$  come within a fraction of a percent of each other. Naturally, rescaling has a larger effect on results obtained with  $\hat{\Sigma}^{(2)}$ . This means that the rescaling really imitates the effect of higher-order correlations and should lead to more accurate results.

TABLE I. Contribution of terms (6)–(8) to the frequencies of the hyperfine transition in the ground state of  $^{133}\text{Cs}$  [ $\delta\nu_0/E^2 \times 10^{-10}$  Hz/(V/m) $^2$ ] in different approximations.

$\hat{\Sigma}$	(6)	(7)	(8)	Total
$\hat{\Sigma}^{(2)a}$	-0.9419	0.0210	-1.0722	-1.9931
$\lambda \hat{\Sigma}^{(2)b}$	-1.0239	0.0229	-1.2688	-2.2697
$\hat{\Sigma}^{(\infty)c}$	-1.0148	0.0232	-1.2706	-2.2622
$\lambda \hat{\Sigma}^{(\infty)b}$	-1.0167	0.0230	-1.2695	-2.2632

<sup>a</sup> $\hat{\Sigma}^{(2)}$  is the second-order correlation potential.

<sup>b</sup>Rescaled  $\hat{\Sigma}$ .

<sup>c</sup> $\hat{\Sigma}^{(\infty)}$  is the all-order correlation potential.

In summary, we have three ways of estimation of the accuracy of calculations: (a) calculation of static polarizability (0.5% accuracy); (b) calculation of the hfs (0.9% accuracy); and (c) comparison of the results obtained in the different most accurate approximations (last three lines in Table I), which differ by about 0.3%. Therefore, we can say that the accuracy of the calculations is not worse than 1%. Our final result is

$$k = -2.26(2) \times 10^{-10} \text{ Hz/(V/m)}^2. \quad (12)$$

This corresponds to  $\beta = -1.70(2) \times 10^{-14}$ . To obtain a frequency shift at finite temperature, one needs to substitute this value into Eq. (3). For accurate results, one also needs to know the value of  $\epsilon$ . It was estimated in Ref. [1] in single-resonance approximation and found to be 0.014. In many-resonance calculation,  $\epsilon = 0.013$  [12].

We present our final result for the frequency shift together with other theoretical and experimental results in Table II. Our value is in good agreement with early measurements [2–4] and *ab initio* calculations [5,6], while recent measurements [7,8] and semiempirical calculations [9–11] give the value which is about 10% smaller. The less accurate measurements of Bauch and Schröder [21] cover both cases. We cannot comment on the disagreement between experimental results. However, the source of disagreement between theoretical results seems to be clear. It comes from the contribution of the continuum spectrum to the summation over the complete set of states in term (6). This term has off-diagonal hfs matrix elements between the ground state and excited states. Since the hfs interaction is

TABLE II. Electrostatic frequency shifts for the hyperfine transition of Cs [ $\delta\nu_0/E^2 \times 10^{-10}$  Hz/(V/m) $^2$ ]; comparison with other calculations and measurements.

This work	Other calculations	Ref.	Measurements	Ref.
-2.26(2)	-1.9(2)	[9]	-2.29(7)	[2]
	-2.2302	[5]	-2.25(5)	[3]
	-2.28	[6]	-2.17(26)	[21]
	-1.97(9)	[10]	-2.271(4)	[4]
	-2.06(1)	[11]	-1.89(12)	[7]
	-2.271(8)	[22]	-2.03(4)	[8]

localized over short distances ( $\sim a_0/Z$ ), it emphasizes the contribution of states with high energies including states in the continuum [since  $\Delta p \Delta x \geq \hbar$ , a small area of localization ( $\Delta x$ ) allows high momentum ( $p$ ) and thus high energy]. In our calculations, the contribution of states above  $7p$  in term (6) is  $-0.35 \times 10^{-1} \text{ Hz}/(\text{V/m})^2$ , which is 15% of the total answer.

In contrast, states above  $7p$  contribute only about 0.05% of the total value of term (8). This is because the summation goes over the matrix elements of the electric dipole operator, which is large on large distances, and thus suppresses the contribution of high-energy states. It is not surprising, therefore, that a semiempirical consideration, involving only discrete spectrum states, gives very good results for the atomic polarizabilities (see, e.g. [10]). However, let us stress once more that the calculation of polarizabilities checks only term (8) and tells us very little about the accuracy of the other two terms, (6) and (7).

The contribution of the states above  $7p$  is even more important for term (7). Their contribution is about 30% of the total value of this term. However, the term itself is small and its accurate treatment is less important.

In *ab initio* calculations by Lee *et al.* [5], the summation over a complete set of states is reduced to solving a radial equation (equations of this type are often called Sternheimer equations after one of the authors of this work). This approach does include the contribution of the continuum spectrum, and the result is in very good agreement with ours (see Table II).

In other *ab initio* calculations by Pal'chikov *et al.* [6], the summation is done via Green functions. This corresponds to a summation over the complete set of states and does include the continuum spectrum. Again, the result is in very good agreement with other *ab initio* calculations (Ref. [5] and the present work).

Recent calculations by Beloy *et al.* in the companion Letter [22] applied a mixed approach, with extensive use of experimental data for lower cesium states and *ab initio* summation over higher states including continuum. The result is in good agreement with fully *ab initio* calculations.

In contrast, the analysis performed in Refs. [9–11] is limited to a discrete spectrum. Adding  $-0.34 \times 10^{-1} \text{ Hz}/(\text{V/m})^2$  [which is the total *tail* contribution from all three terms (6)–(8) found in our calculation] to the results of Feitchner *et al.* [9] and Micalizio *et al.* [10] brings them to excellent agreement with *ab initio* calcu-

lations. The same modification of the result by Ulzega *et al.* [11] makes it a little bit too large but still closer to other results than without the *tail* contribution.

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