Ytterbium in Quantum Gases and Atomic Clocks: van der Waals Interactions and Blackbody Shifts

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Ytterbium in quantum gases and atomic clocks: van der Waals interactions and blackbody shifts

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We evaluated the C_6 coefficients of Yb-Yb and Yb -alkali/group II van der Waals interactions with 2% uncertainty. The only existing experimental result for such quantities is for the Yb-Yb dimer. Our value, $C_6 = 1929(39)$ a.u., is in excellent agreement with the recent experimental determination of 1932(35) a.u. We have also developed a new approach for the calculation of the dynamic correction to the blackbody radiation shift. We have calculated this quantity for the Yb $6s^2\ ^1\mathrm{S}_0 - 6s6p\ ^3\mathrm{P}^o_0$ clock transition with 3.5% uncertainty. This reduces the fractional uncertainty due to the blackbody radiation shift in the Yb optical clock at 300 K to 10^{-18} level.

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I. INTRODUCTION

Ytterbium (Yb: Z=70) has recently emerged as a subject of great interest in ultracold chemistry, physics, and metrology. The observation of state-resolved ionic chemical reactions was recently reported for the $\mathrm{Yb}^+ + \mathrm{Rb} \longrightarrow \mathrm{Yb} + \mathrm{Rb}^+$ system [1]. Yb is a favorite candidate for the studies of ultracold gas mixtures. For example, Li and Yb mixtures have recently been brought to simultaneous quantum degeneracy [2–5] and it appears that cold YbLi molecules could be formed by magnetoassociation [6]. Controlled production of ultracold YbRb* molecules by photoassociation in a mixture of Rb and Yb gases was recently reported in [7, 8]. Such mixtures are of interest for producing ultracold polar molecules for study of dipolar quantum matter, fundamental symmetry studies, and many-body quantum simulation [5]. The availability of five bosonic and two fermionic stable isotopes makes Yb especially attractive for studies of multicomponent superfluids.

The spectrum of Yb contains a number of long-lived excited states that are conveniently accessed by optical techniques. This makes Yb an excellent candidate for atomic parity violation (APV) studies that test the Standard Model of electroweak interactions, put limits on its possible extensions, constrain parameters of weak hadronic interactions, and may yield information on neutron distributions within nuclei [9, 10]. The APV signal recently observed in the Yb $6s^2\ ^1\mathrm{S}_0 - 5d6s\ ^3\mathrm{D}_1$ 408-nm forbidden transition [10, 11] is two orders of magnitude larger than in Cs, subject of the most accurate APV study to date. Such long-lived states are also convenient for the development of next-generation ultra-precise frequency standards. The Yb $^1\mathrm{S}_0 \rightarrow {}^3\mathrm{P}^o_0$ 578-nm transition now provides one of the world's most accurate optical atomic frequency standards [12, 13].

The work carried out in this Letter is pertinent to all applications mentioned above. Our two main subjects are the determination of van der Waals C_6 coefficients that characterize the long-range interactions between two atoms, and the blackbody radiation (BBR) shifts of the two states in atomic clock transitions. Knowledge of the long range interactions in Yb-Yb and Yb-alkali/group II dimers is critical to understanding the physics of dilute gas mixtures. The dynamic correction to the BBR shift is one of the largest irreducible contributions to uncertainty budget of the Yb clock [12], and it is difficult to measure directly. These two seemingly disparate topics both require accurate determination of frequency-dependent atomic polarizabilities over a wide range of frequencies. Therefore, it is natural to consider them in the same work. A future accurate theoretical determination of the APV amplitude in Yb requires a similar approach, and this work provides a background for such studies.

We carry out the calculation of frequency-dependent atomic polarizabilities using the first-principles approach that combines configuration interaction (CI) with the coupled-cluster all-order approach (CI+all-order) that treats both core and valence correlation to all orders. Several new method developments are presented in this work. First, we have implemented the reduced linear equation (RLE) and direct inversion in iterative subspace (DIIS) stabilizer procedures described in Ref. [14] into the coupled-cluster part of the CI+all-order method. Otherwise, the construction of the effective Hamiltonian needed for the incorporation of the core and core-valence correlations into the CI method could not be carried out due to convergence problems associated with extremely large correlations involving the 4f shell. Second, we have applied the CI+all-order method for the first time to the calculation of C_6 coefficients. Finally, we have developed a new approach to the calculation of the dynamic correction to the blackbody radiation shift in terms of the second partial derivative with respect to frequency of the dynamic polarizability, as obtained from the solution of the inhomogeneous equation in the valence sector. Previous calculations of the dynamic correction to the BBR shift accounted for the contributions of just a few intermediate states to the polarizability [15].

Unless stated otherwise, we use atomic units (a.u.) for all matrix elements and polarizabilities throughout this paper. The atomic unit for polarizability can be converted to SI units via α/h [Hz/(V/m)^2]=$2.48832\times10^{-8}\alpha$ (a.u.), where the conversion coefficient is $4\pi\epsilon_0 a_0^3/h$, a_0 is the Bohr radius and ϵ_0 is the electric constant.

Calculation of Yb properties requires an accurate treatment of both core-valence and valence-valence correlations. This can be accomplished within the framework of the CI+all-order method that combines configuration interaction and coupled-cluster approaches [16–18]. Here we report the extension of this method that resolves the convergence problems associated with particularly large correlation corrections as well as apply it for the first time to the calculation of the C_6 coefficients. We refer the reader to Refs. [16, 17] for detailed description of this approach, and here we report only new method developments specific to this work. In order to establish the accuracy of our approach, we also perform the pure CI and the CI combined with many-body perturbation theory (CI+MBPT) calculations carried out with the same parameters such as basis set, configuration space, number of partial waves, etc..

The single-electron energies and the wave functions are found from the solution of the Dirac-Hartree-Fock (DHF) equations. Then the wave functions and the low-lying energy levels are determined by solving the multiparticle relativistic equation for two valence electrons [19], $H_{\rm eff}(E_n)\Phi_n = E_n\Phi_n$. The effective Hamiltonian is defined as $H_{\rm eff}(E) = H_{\rm FC} + \Sigma(E)$, where $H_{\rm FC}$ is the Hamiltonian in the frozen-core approximation. The energy-dependent operator $\Sigma(E)$ which takes into account virtual core excitations is constructed using second-order perturbation theory in the CI+MBPT method [20] and using a linearized coupled-cluster single-double method in the CI+all-order approach [16]. However, the CI+all-order approach developed in [16, 17] could not be directly implemented for Yb owing to convergence problems of the all-order equations associated with large oscillations of the iterative solution due to very large correlations in the $4f$ shell. Both the Yb^{2+} core and some of the Yb$^+$ valence shell all-order equations that are used to construct the effective Hamiltonian diverge using conventional iteration schemes. We have resolved this problem by using RLE and DIIS convergence stabilizers described in [14] within the framework of the CI+all-order method. Both the DIIS and RLE methods seek to minimize the error between the iteratively found solutions of the all-order equations and the exact answer using a least-squares approach to the error minimization. Since the exact answer is unknown, the approximate solution is constructed as a linear combination of a series of iteratively found solutions.

We present the energy levels obtained in the CI, CI+MBPT, and CI+all-order approximations and compare them with the experimental values [21] in Table I of the supplementary material [22]. At the CI stage, the theoretical energy levels differ rather significantly from the experimental energies, up to 19% for the $6s6p$ states. Including the core-valence correlations in the second order of the MBPT improves the agreement to the 1.5-5.5% level. Further improvement of the theoretical energies is achieved when the CI+all-order approximation is used. The two-electron binding energy of the ground state is accurate to 0.7% with the *ab initio* CI+all-order approach, a factor of 2 improvement in comparison with the CI+MBPT result.

The valence part of the polarizability is determined by solving the inhomogeneous equation of perturbation theory in the valence space, which is approximated as

$$(E_v - H_{\rm eff})|\Psi(v,M')\rangle = D_{{\rm eff},q}|\Psi_0(v,J,M)\rangle \qquad (1)$$

for a valence state v with the total angular momentum J and projection M [23]. The effective dipole operator $D_{\rm eff}$ includes random phase approximation (RPA) corrections. The ionic core part of the polarizability, α_c, is calculated separately in the RPA and is found to be $\alpha_c = 6.4$ a.u. The small valence-core (vc) α_{vc} term that corrects the ionic core polarizability for the presence of the valence electrons is also calculated in the RPA; it is equal to -0.4 a.u. and -0.2 a.u. for the $6s^2\ ^1\mathrm{S}_0$ and $6s6p\ ^3\mathrm{P}_0$ states, respectively. DHF calculations are carried out as well for both of these contributions to evaluate the uncertainty associated with these terms, which was found to be negligible at the present level of accuracy. The contributions to the $6s^2\ ^1\mathrm{S}_0$ and $6s6p\ ^3\mathrm{P}^o_0$ polarizabilities α_0 of Yb are given in Table II of the supplementary material [22].

Accurate calculation of the polarizabilities of low-lying states is more difficult for Yb than for alkaline-earth atoms. It is known that the main contribution to the ground state polarizability of Yb comes from $4f^{14}6s6p\ ^1\mathrm{P}^o_1$ and $4f^{13}5d6s^2\,(7/2,5/2)^o_1$ states (see, e.g., [24]). The energy difference between these states is only 3789 cm^{-1} and they strongly interact with each other. Fig. 1 of the supplementary material illustrates positions of the relevant energy levels. Calculations that treat Yb as an atom with only two valence electrons fail to account properly for the interaction between valence and core-excited states and describe states with an unfilled f shell. While the state $4f^{13}5d6s^2\,(7/2,5/2)^o_1$ does not belong to the valence subspace and is not directly mixed with the $4f^{14}6s6p\ ^1\mathrm{P}^o_1$ state in our calculations, its effect is introduced via the calculation of the effective Hamiltonian, since we allow all single and double excitations of the core shells during its construction. As a result, the polarizability calculation carried out via the solution of the inhomogeneous equation does not appear to be affected by this problem. A theoretical explanation of this fact was suggested in [25] which considered mixed and unmixed basis sets that included $4f^{14}6s6p\ ^1P^o_1$ and $4f^{13}5d6s^2\,(7/2,5/2)^o_1$ states. Excellent agreement of our results with all measured Yb polarizability-related prop-

TABLE I: The $6s^2\,{}^1\mathrm{S}_0$ and $6s6p\,{}^3\mathrm{P}^o_0$ static polarizabilities $\alpha_g(\omega=0)$ of Yb and their difference $\Delta\alpha \equiv \alpha({}^3\mathrm{P}^o_0) - \alpha({}^1\mathrm{S}_0)$ calculated in CI, CI+MBPT, and CI+all-order approximations in a.u. The CI+all-order values are taken as final. The present results are compared with other theoretical and experimental values.

Method	$\alpha({}^1\mathrm{S}_0)$	$\alpha({}^3\mathrm{P}^o_0)$	$\Delta\alpha$
CI	187.9	279.7	91.6
CI+RPA	166.1	258.4	92.3
CI+MBPT+RPA	138.3	305.9	167.5
CI+all-order+RPA	140.9	293.2	152.3
Final	141(2)	293(10)	152
Theory [15] (2006)	111.3(5)	266(15)	155
Theory [24] (2007)	143		
Theory [26] (2008)	144.6		
Theory [25] (2010)	141(6)	302(14)	161
Ref. [27]^a (2012)	134.4 − 144.2	280 − 290	
Expt. [13] (2012)			145.726(3)

^{a}Constraints based on experimental data. The uncertainty in each of these values is 1.0.

erties, including Stark shift and magic wavelength of the $6s^2\,{}^1\mathrm{S}_0 - 6s6p\,{}^3\mathrm{P}^o_0$ transition and the C_6 coefficient of the Yb-Yb dimer, confirms that the mixing problem does not appear to affect such properties. We note that this is only true as long as no experimental data is substituted for theoretical CI+all-order quantities in any part of the calculations, since this will compromise the basis set completeness [25]. It follows that the direct solution of the inhomogeneous equation may be more accurate than expected from the comparison of the individual matrix elements with experiment. This conclusion is important for future calculation of the parity-violating amplitudes that could by evaluated by the same techniques.

Table I presents results for the static polarizabilities of the $6s^2\,{}^1S_0$ and $6s6p\,{}^3P^o_0$ states and their differences. We note that the states with an unfilled $4f$ shell contributed less to the polarizability of the ${}^3\mathrm{P}^o_0$ than ${}^1\mathrm{S}_0$ state. In particular, even-parity states with an unfilled $4f$ shell lie rather high in energy and their contributions to the polarizability and influence on other states is not so significant. The results obtained in the CI, CI+RPA, CI+MBPT+RPA, and CI+all-order+RPA approximations are presented. Our recommended values obtained at the CI+all-order+RPA stage are in a reasonable agreement with other theoretical values. We emphasize that our calculations are completely *ab initio*. The most recent recommended values of Ref. [25] include adjustment to reproduce the experimental value of the magic wavelength. The set of accurate experimental data was used to set upper and lower bounds on the ${}^1\mathrm{S}_0$ and ${}^3\mathrm{P}^o_0$ polarizabilities in [27]. Our recommended values are in excellent agreement with these constraints taking an account the uncertainties. We can roughly estimate the uncertainty of our calculations as the difference of the CI+MBPT and CI+all-order values, which yields 1.8% and 4.3% for ${}^1\mathrm{S}_0$ and ${}^3\mathrm{P}^o_0$ states. We note that the CI+all-order value is higher than CI+MBPT for ${}^1\mathrm{S}_0$ but lower for ${}^3\mathrm{P}^o_0$, so we can expect that these uncertainties will add cumulatively for the $\Delta\alpha$ polarizability difference. However, our value of $\Delta\alpha$ agrees with a recent experiment to 4.3%, so our values are somewhat more accurate than the estimates above (1% and 3.5%, respectively). A direct measurement of the ground state polarizability with 1% accuracy would be an excellent test of the quality of calculations.

To further check the accuracy of our approach we calculated the magic wavelength λ for the ${}^1\mathrm{S}_0$ and ${}^3\mathrm{P}^o_0$ states. At the magic wavelength, the frequency-dependent polarizabilities of the two states are equal. We obtain $\lambda = 754$ nm in the CI+all-order approximation which is within 1% of the experimental value 759.355 nm [28]. The polarizability of the ${}^3\mathrm{P}^o_0$ state grows rapidly in the vicinity of the intersection of the ac polarizabilities. It means that even a small change in λ leads to a significant change in $\alpha({}^3\mathrm{P}^o_0)$. For example, the CI+MBPT value is significantly higher, 789 nm. Such close agreement of the CI+all-order value with the experimental wavelength confirms the accuracy of the polarizabilities quoted above.

An important application of the polarizability calculation is to determine the shift of the ${}^1\mathrm{S}_0 - {}^3\mathrm{P}^o_0$ transition frequency by the effects of the ambient blackbody radiation. The BBR shift is now one of the largest irreducible contributions to the budget of the uncertainty of optical atomic clocks. The leading contribution to the BBR shift of the energy level g can be expressed in terms of its static polarizability $\alpha_g(\omega=0)$ by [15]

$$\Delta E_g = -\frac{2}{15}(\alpha\pi)^3(k_BT)^4\alpha_g(0)\left[1+\eta\right], \qquad (2)$$

where k_B is the Boltzmann constant, T is the temperature, and η is a "dynamic" fractional correction to the total shift that reflects the averaging of the frequency dependence polarizability over the frequency of the blackbody radiation spectrum.

The dynamic correction η can be be approximated by

$$\begin{aligned}\eta &= \eta_1+\eta_2+\eta_3 = \frac{80}{63\,(2J_g+1)}\,\frac{\pi^2}{\alpha_g(0)k_BT} \\ &\times \sum_n \frac{|\langle n||D_{\mathrm{eff}}||g\rangle|^2}{y_n^3}\left(1+\frac{21\pi^2}{5\,y_n^2}+\frac{336\pi^4}{11y_n^4}\right), \quad (3)\end{aligned}$$

where $y_n = (E_n - E_g)/(k_BT)$ [15]. We express the dominant term in the equation above as the second derivative of the polarizability:

$$\eta_1 = \frac{20}{21\,(2J_g+1)}\,\frac{(\pi k_BT)^2}{\alpha_g(0)}\,\frac{\partial^2}{\partial E_g^2}\alpha_g(0) \qquad (4)$$

and find $\eta_1({}^1\mathrm{S}_0) = 0.00116$ and $\eta_1({}^3\mathrm{P}_0) = 0.00934$. We calculated the second term in Eq. (3) using both a forth derivative of α and sum over states with the CI+all-order values of the matrix elements; identical result $\eta_2({}^3\mathrm{P}_0) = 0.00029$ was obtained. η_2 is negligible for ${}^1\mathrm{S}_0$, 0.000003. The third term can be neglected at the present level of

TABLE II: The values of the C_6 coefficients (in a.u.) for the homonuclear Yb dimer and the heteronuclear alkali-metal/group II - Yb dimers. All atoms are in their ground states. The $\alpha(i\omega)$ for alkali and Mg, Sr, and Ca are taken from Ref. [29] in rows CI+MBPT and CI+all$^{(a)}$. The $\alpha(i\omega)$ for Mg and Ca are calculated with the CI+all-order method in the present work in row CI+all$^{(b)}$. The present CI+MBPT and CI+all-order Yb $\alpha(i\omega)$ values are used in rows CI+MBPT and CI+all-order$^{(a,b)}$, respectively. The uncertainty of the final CI+all-order values is estimated to be 2%.

	Li-Yb	Na-Yb	K-Yb	Rb-Yb	Cs-Yb
CI+MBPT	1534	1655	2548	2807	3367
CI+all$^{(a)}$	1551	1672	2576	2837	3403
Theory [30]	1594				
	Yb-Yb	Mg-Yb	Ca-Yb	Sr-Yb	
CI+MBPT	1901	1086	2000	2414	
CI+all$^{(a)}$		1093	2017	2435	
CI+all$^{(b)}$	1929	1092	2024		
Expt. [31]	1932(35)				

accuracy. The resulting values of the dynamic corrections to the BBR shift at 300 K are $\Delta\nu_{\rm BBR}(^1{\rm S}_0) = -0.0014$ Hz and $\Delta\nu_{\rm BBR}(^3{\rm P}_0) = -0.0243$ Hz, respectively.

The total dynamic correction to the BBR shift at 300 K is determined as the difference between the individual shifts, $\Delta\nu^{\rm dyn}_{\rm BBR} = -0.0229(8)$ Hz. The uncertainty is taken to be 3.5% based on the uncertainty in the CI+all-order value of the ^{3}P$_0$ polarizability obtained above. Combining this result with the experimental determination of the $\Delta\alpha = 145.726(3)$ a.u. that yields $\Delta\nu^{\rm static}_{\rm BBR} = -1.25484(3)$ Hz [13], we get the final result for the BBR shift at 300 K: $\Delta\nu_{\rm BBR} = -1.2777(8)$ Hz. This value is in excellent agrement with the determination of the BBR shift mostly from the experimental data $\Delta\nu_{\rm BBR} = -1.2774(6)$ Hz that was just reported in [32]. Details of the calculation of the dynamic correction to the BBR shift are given in the supplementary material [22].

Many of the same considerations concerning accurate calculation of the frequency-dependent polarizability arise in the calculation of the van der Waals coefficients. If two atoms A and B have spherically symmetrical ground states, the leading power of the long-range interactions takes the form $V(R) = -C_6^{AB}/R^6$, where R is the distance between atomic nuclei. The the van der Waals coefficient C_6^{AB} can be calculated as [33]

$$C_6^{AB} = \frac{3}{\pi}\int_0^\infty \alpha^A(i\omega)\,\alpha^B(i\omega)d\omega, \qquad (5)$$

where $\alpha(i\omega)$ is the frequency-dependent polarizability at an imaginary frequency. In practice, we compute the C_6^{AB} coefficients by approximating the integral (5) by Gaussian quadrature of the integrand computed on the finite grid of discrete imaginary frequencies [34].

For the alkali and group II atoms, we use frequencies and weights tabulated in Ref. [29] at 50 points. These dynamic polarizabilities were obtained by combining high-precision experimental data for matrix elements of principal transitions with high-precision many-body methods, such as linearized coupled-cluster approach and CI+MBPT. The accuracy of the corresponding homonucelar C_6 was estimated to be better than 1% for all cases relevant in this work with the exception of Ca, were it was 1.5% [29].

The Yb imaginary frequency polarizabilities $\alpha(i\omega)$ for the ground ^{1}S$_0$ state are calculated in this work by solving the inhomogeneous equation (1) with the appropriate modifications. We use the same 50-point frequency grid as in Ref. [29] for consistency. To evaluate the uncertainty in the C_6 coefficients, we carried out both CI+MBPT and CI+all-order calculations of the ground state $\alpha(i\omega)$. The same alkali and group II data are used in both cases. The results are summarized in Table II. We find that the differences between CI+MBPT and CI+all-order results are actually smaller (1-1.5%) than for the ground state static polarizability (1.8%) since the differences decrease with ω for $\alpha(i\omega)$ for Yb. As a result, we expect the accuracy of the C_6 to be on the same order as the static polarizability, rather than larger by a factor of two. Moreover, our value for the C_6 coefficient of the homonuclear Yb dimer is in excellent agreement with the experimental result [31], which is accurate to 1.8%. Comparison of the present value of the C_6 coefficient for the homonuclear Yb dimer with other theoretical results is given in Table II of the supplementary material [22]. Our Li-Yb value is consistent with coupled-cluster value from [30] within the uncertainties. Based on the comparison of the CI+MBPT and CI+all-order values for heteronuclear C_6 coefficients, and agreement with experiment for the Yb ^{3}P$_0-^1$S$_0$ Stark shift and magic wavelengths, and C_6 coefficient for Yb-Yb dimer, we estimate that our predictions of the C_6 coefficients for the heteronuclear alkali-metal atom/group II - Yb dimers are accurate to about 2%.

In conclusion, we have carried out fully *ab initio* all-order calculations of Yb properties. Our values of the Yb ^{1}S$_0-{}^3$P$^o_0$ Stark shift and magic wavelength as well as the C_6 coefficient of the Yb$_2$ dimer are in excellent agreement with experiment. We have developed a new approach of calculation of the dynamic correction to the BBR shift that does not involve an explicit sum over states. The Stark shift of the clock transition was determined experimentally [13] with a high precision. As a result, the uncertainty in the dynamic correction can now be directly related to the uncertainty of the BBR shift of this transition. Thus, when combined with the recent measurement of the Yb clock Stark shift [13], our calculation of the dynamic correction allows us to reduce the fractional uncertainty due to the BBR shift in the Yb optical lattice clock to 10^{-18} level. The same method can be used to evaluate the dynamic correction for any optical atomic clock. Finally, we have presented the first recommended values of C_6 coefficients for alkali/group II-Yb dimers for future experimental efforts in producing ultracold polar molecules.

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