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Determination of lifetimes of $6P_J$ levels and ground-state polarizability of Cs from the van der Waals coefficient C_6

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A method for determination of atomic lifetimes from the value of dispersion coefficient C_6 of molecular potentials correlating to two ground-state atoms is proposed. The method is illustrated with atomic Cs using C_6 deduced from high-resolution Feshbach spectroscopy. The following lifetimes are determined: $\tau_{6P_{1/2}}=34.80\pm0.07$ ns and $\tau_{6P_{3/2}}=30.39\pm0.06$ ns. The underlying electric-dipole matrix elements are required for a refined analysis of parity violation in ^{133}Cs. We also obtain static polarizability of the Cs ground state, $\alpha(0)=400.49(81)$ atomic units. It is demonstrated that the uncertainties may be considerably reduced as the coefficient C_6 is constrained further.

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The leading long-range interaction of two atoms in their respective spherically symmetric ground states is described in terms of van der Waals coefficient C_6 [1]. Studies of magnetic-field induced Feshbach resonances and photoassociation in ultracold atomic samples allow us to constrain this coefficient. Several highly accurate determinations of C_6 for alkali-metal dimers were recently reported [2–4]. Here we propose a method for determination of atomic lifetimes from these coefficients.

The method is illustrated with atomic Cs. A renewed attention has been given to the structure of this atom over the last few years following the most accurate to date measurement [5] of atomic parity nonconservation (PNC). Atomic PNC [6] provides powerful constraints on possible ‘‘new physics’’ beyond the standard model of elementary particles. At present the dominant theoretical atomic-structure uncertainty precludes an improved interpretation of the measurement of parity violation. Major theoretical efforts so far have focused on Breit [7], neutron skin/halo [8], and radiative corrections [9]. The latest analyses [9], depending on the estimate of theoretical accuracy, report a disagreement with a prediction of standard model as large as 2.2 standard deviations, hinting at a possible evidence for an extra Z boson.

To refine the interpretation of parity violation in ^{133}Cs, it is crucial to extend the previous *ab initio* relativistic many-body calculations [10] to higher orders in residual Coulomb interaction. In this context, high-precision lifetime measurements serve a vital function in assessing the role of omitted contributions. In addition, the underlying electric-dipole matrix element for the $6\,P_{1/2}\rightarrow 6\,S_{1/2}$ transition sensitively enters the expression for the parity-violating $6S_{1/2}$-$7S_{1/2}$ amplitude [11]. There were two previous high-accuracy direct measurements of lifetimes of $6P_{1/2,3/2}$ levels in Cs by Young *et al.* [12] and by Rafac *et al.* [13]. While the precision of these experiments is similar, the measured lifetimes of the $6\,P_{1/2}$ level $\tau_{6\,P_{1/2}}$ differ by more than three standard deviations. Our high-precision indirect determination results in $\tau_{6\,P_{1/2}}$ in close agreement with the measurements [12], providing an independent reference for needed refined calculations of parity violation.

In recent years the most accurate lifetimes for alkali-metal atoms were derived using photoassociation spectroscopy of ultracold atomic samples reviewed in Refs. [14]. Unfortunately, this approach was not as successful for Cs because of peculiarities of molecular potentials of the Cs dimer [15]. Based on the van der Waals coefficient C_6 deduced from ultracold collision data, the calculations reported here lead to the matrix elements for Cs with an uncertainty similar to the best direct lifetime measurements. For Cs, Leo *et al.* [3] deduced $C_6=6890(35)$ a.u. using results of high-resolution Feshbach spectroscopy. [4] Recently more Feshbach resonances due to higher angular momenta were identified and this value has been constrained further to $C_6=6859(25)$ au. [16].

I. OVERVIEW AND RESULTS

The dispersion coefficient C_6 may be expressed as a quadrature of dynamic polarizability [1] (unless specified otherwise, atomic units $\hbar=|e|=m_e=1$ are used throughout):

$$C_6=\frac{3}{\pi}\int_0^\infty d\omega\,[\alpha(i\omega)]^2, \tag{1}$$

where

$$\alpha(i\omega)=\frac{2}{3}\sum_{|i\rangle}\frac{\Delta E_i}{(\Delta E_i)^2+\omega^2}|\langle v|\mathbf{D}|i\rangle|^2. \tag{2}$$

Here $|v\rangle$ is the atomic ground state ($6S_{1/2}$ for Cs), ΔE_i are energies of intermediate states $|i\rangle$ taken with respect to the ground state, and $\mathbf{D}$ is an electric-dipole operator. For Cs almost 85% of C_6 is accumulated from intermediate states $6P_{1/2}$ and $6P_{3/2}$. We exploit this strong dependence to deduce matrix elements of principal transitions by calculating residual contributions using *ab initio* methods. We separate the contribution of $6P_J$ states to dynamic polarizability and write

$$\alpha(i\omega)=\alpha_p(i\omega)+\alpha_r(i\omega),$$

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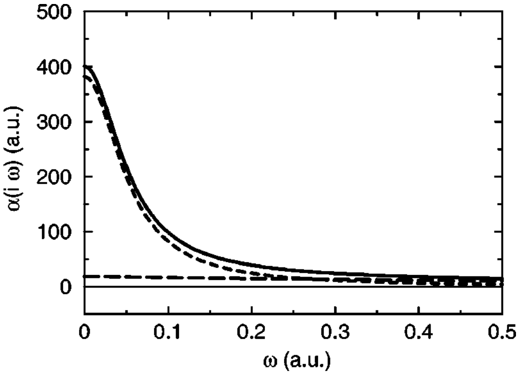

FIG. 1. Comparison of contributions to dynamic dipole polarizability from the principal transitions $\alpha_p(i\omega)$ (dashed line) and residual intermediate states $\alpha_r(i\omega)$ (long-dashed line). Total polarizability $\alpha(i\omega)$ is represented by a solid line.

where α_r combines contributions of other intermediate states. These polarizabilities are shown in Fig. 1. Introducing reduced matrix elements $D_J = \langle 6P_J || D || 6S_{1/2} \rangle$ and a ratio $R = (D_{3/2}/D_{1/2})^2$,

$$\alpha_p(i\omega) = \frac{D_{1/2}^2}{3}\left(\frac{\Delta E_{1/2}}{\Delta E_{1/2}^2 + \omega^2} + \frac{\Delta E_{3/2}}{\Delta E_{3/2}^2 + \omega^2} R \right). \qquad (3)$$

Energies ΔE_J of $6P_J$ states are known experimentally with a high accuracy and the ratio of matrix elements $R = 1.9809(9)$ has been measured with a 0.05% precision [17]. The atomic lifetimes τ_{6P_J} may be expressed as $\tau_{6P_J}^{-1} = 2.1421 \times 10^{10} \Delta E_J^3 D_J^2/(2J+1)\,\mathrm{s}^{-1}$.

Since

$$C_6 = \frac{3}{\pi}\int_0^\infty d\omega [\alpha_p(i\omega)^2 + 2\alpha_p(i\omega)\alpha_r(i\omega) + \alpha_r(i\omega)^2], \qquad (4)$$

the van der Waals coefficient can be parametrized in terms of the matrix element $D_{1/2}$ as

$$C_6 = D_{1/2}^4 \xi_p + D_{1/2}^2 \xi_x + \xi_r, \qquad (5)$$

where

$$\xi_p = \frac{1}{12}\left(\frac{1}{\Delta E_{1/2}} + \frac{4R}{\Delta E_{1/2} + \Delta E_{3/2}} + \frac{R^2}{\Delta E_{3/2}} \right), \qquad (6)$$

$$\xi_x = \frac{2}{\pi}\int_0^\infty d\omega\, \alpha_r(i\omega) \left(\frac{\Delta E_{1/2}}{\Delta E_{1/2}^2 + \omega^2} + \frac{\Delta E_{3/2}}{\Delta E_{3/2}^2 + \omega^2} R \right), \qquad (7)$$

$$\xi_r = \frac{3}{\pi}\int_0^\infty d\omega [\alpha_r(i\omega)]^2. \qquad (8)$$

Solving the quadratic equation (5) we obtain

$$D_{1/2}^2 = \sqrt{\left(\frac{C_6 - \xi_r}{\xi_p}\right) + \left(\frac{\xi_x}{2\xi_p}\right)^2} - \frac{\xi_x}{2\xi_p} \qquad (9)$$

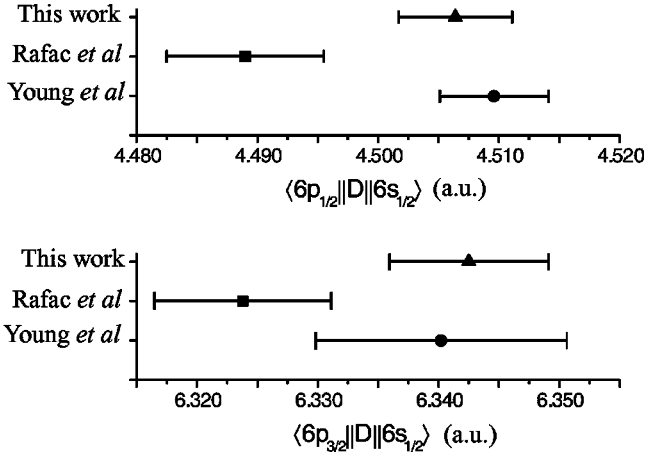

FIG. 2. Comparison of deduced values for matrix elements $\langle 6P_{1/2} || D || 6S_{1/2} \rangle$ (top panel) and $\langle 6P_{3/2} || D || 6S_{1/2} \rangle$ (bottom panel) with the results of direct lifetime measurements by Rafac *et al.* [13] and by Young *et al.* [12].

and the problem is reduced to an accurate determination of parameters ξ. The calculation of these quantities and uncertainty estimates are discussed below. We find $\xi_p = 14.0787(84)$, $\xi_x = 45.12(30)$, and $\xi_r = 136.7(2.7)$ and obtain

$$\langle 6P_{1/2} || D || 6S_{1/2} \rangle = 4.5064(47), \qquad (10)$$

$$\langle 6P_{3/2} || D || 6S_{1/2} \rangle = 6.3425(66). \qquad (11)$$

The comparison of the dipole matrix elements with the results of previous direct high-precision lifetime measurements [12,13] is presented in Fig. 2. The accuracy of our method is comparable to that of these measurements. One notes a large discrepancy between results of the previous determinations for the important matrix element $\langle 6P_{1/2} || D || 6S_{1/2} \rangle$ entering the parity-violating amplitude. Our values support experimental results by Young *et al.* [12].

II. DETAILS OF CALCULATION

The parameter ξ_p can be evaluated with a high precision using experimental energies and the ratio of matrix elements R from Ref. [17]. We obtain $\xi_p = 14.0787(84)$; the uncertainty is induced by experimental error in the ratio R.

To determine parameters ξ_x and ξ_r we have to compute the residual dynamic polarizability $\alpha_r(i\omega)$. Here we follow formalism laid out in Ref. [18] and augment it with a refined error analysis. The intermediate states can be separated into two classes-valence states, both bound and continuum (α_v'), and core-excited states (α_c)

$$\alpha_r(i\omega) = \alpha_v'(i\omega) + \alpha_c(i\omega) + \alpha_{cv}(i\omega).$$

Here the term α_{cv} contains a small core-valence coupling correction addressed below.

To sum over valence states α_v' we use the Dalgarno-Lewis method restricted to the valence space. We computed the full valence polarizability by solving Dalgarno-Lewis inhomoge-

TABLE I. Breakdown of various *ab initio* contributions to static dipole polarizability (in a.u.) from the intermediate states beyond $6P_J$.

	Value	Method
$\alpha'_v(0)$ $(7\cdots\infty P_J)$	1.81	MBPT
$\alpha_{cv}(0)$	-0.72	RPA
$\alpha_c(0)$	15.81	RPA
$\alpha_r(0)$	16.91	
$\alpha_r^{\rm se}(0)$	16.74(11)	

neous equation and subtracted explicit contribution of principal transitions obtained at the same level of many-body perturbation theory. Correlations were accounted for with the relativistic many-body perturbation theory; we included random-phase approximation (RPA) and self-energy contributions in the computations. In the summation over core-excited states we employ relativistic random-phase approximation [19] generalized for frequency dependence. In the RPA we allow for excitations of core electrons to all possible valence states, including the occupied $6S$ state. To account for a subsequent violation of the Pauli exclusion principle, a counter core-valence coupling term α_{cv} was introduced.

We calculate the parameter ξ_x with the quadrature (7). We notice that in Eq. (7) a smooth and broad curve $\alpha_r(i\omega)$ is integrated with a narrow Lorentzian-shaped function $\alpha_p(i\omega)$ (see Fig. 1). To illustrate the main source of uncertainty we approximate

$$\xi_x \approx \alpha_r(0)\,\frac{6}{\pi}\int_0^\infty \frac{\alpha_p(i\omega)}{D_{1/2}^2}\,d\omega = \alpha_r(0)(1+R), \tag{12}$$

i.e., the uncertainty in ξ_x is governed by static residual polarizability $\alpha_r(0)$. We present a breakdown of various contributions to this polarizability in Table I.

The error bars of the derived matrix elements depend sensitively on the uncertainty of ξ_x. As a test case we consider contribution of principal transitions to the static dipole polarizability

$$\alpha_p(0) = \frac{1}{3}\left(\frac{D_{1/2}^2}{\Delta E_{1/2}} + \frac{D_{3/2}^2}{\Delta E_{1/2}}\right). \tag{13}$$

With the employed many-body method we obtain $\alpha_p(0) = 383.6$, while the corresponding semiempirical values, calculated with the experimental energies and matrix elements [12,13] are 383.9(1.1) and 381.25(96), respectively. Based on this test case we assign a conservative 1% uncertainty to the $\alpha'_v(0)$ contribution. The core-valence coupling term, $\alpha_{cv}(i\omega)$, was estimated as a half of the difference between two determinations of core polarizability—in the first case we explicitly exclude core excitations to the 6s shell, and in the second case these excitations are allowed. For the $\alpha_{cv}(0)$ we obtain -0.47 in the Dirac-Hartree-Fock (DHF) approximation and -0.72 in more sophisticated RPA method. We assign a 15% error bar to the RPA value estimated as a half of the difference between the DHF and RPA results for $\alpha_{cv}(0)$. Finally the core polarizability $\alpha_c(0)$ was calculated in the RPA approximation. To improve the overall accuracy, we further replace the *ab initio* $\alpha_c(0)$ with semiempirical value 15.644(5) [20] and obtain $\alpha_r^{\rm se}(0) = 16.74(11)$. It is worth noting that the overall accuracy of the $\alpha_r(0)$ and the derived matrix elements may be further improved using, for example, linear-response coupled-cluster method [21]. Based on Eq. (12) we improve the accuracy of the parameter ξ_x by rescaling the *ab initio* value of the integral, Eq. (7), with the semiempirical $\alpha_r^{\rm se}(0)$. The final value for the quantity ξ_x is 45.12(30).

The overwhelming contribution to parameter ξ_r comes from the dynamic core polarizability $\alpha_c(i\omega)$. The calculated RPA dynamic polarizability of the core satisfies an important Thomas-Reiche-Kuhn (TRK) sum rule $\lim_{\omega\to\infty}\alpha_c(i\omega) = N_c/\omega^2$, N_c being the number of core electrons ($N_c = 54$ for Cs.) The static ($\omega = 0$) core polarizability obtained with RPA is $\alpha_c(0) = 15.81$. This result is in a 1% agreement with a value of 15.644(5) deduced from semiempirical fitting of Rydberg spectrum of Cs [20]. The RPA accounts only for a correlated contribution of particle-hole excitations. However, the contribution from multiple core excitations is expected to be strongly suppressed because they may couple only to the first-and higher-order many-body corrections to the core wave function. In addition, their effect is reduced by larger energy denominators in the expression for polarizability.

Overall the RPA approximation results in a dynamic core polarizability differing from the high-precision value by 1% at $\omega = 0$ and at large frequencies recovering the exact TRK limit [22]. The parameter ξ_r calculated including only core polarizability, $\xi_r = 137.2$, is slightly decreased by inclusion of valence states and the core-valence coupling term α_{cv} to 136.7. Based on the above discussion we assign a 2% uncertainty to ξ_r and arrive at $\xi_r = 136.7(2.7)$.

Employing $C_6 = 6859(25)$ [16] and the calculated parameters ξ we determine the matrix element $D_{1/2}$, and, using the ratio R [17], the matrix element $D_{3/2}$. The obtained values are given in Eq. (10). The uncertainty in matrix element calculated from Eq. (9) may be parametrized as

$$\left(\frac{\delta D_{1/2}}{D_{1/2}}\right)^2 = A_{C_6}\left(\frac{\delta C_6}{C_6}\right)^2 + A_x\left(\frac{\delta \xi_x}{\xi_x}\right)^2 + A_r\left(\frac{\delta \xi_r}{\xi_r}\right)^2 + A_R\left(\frac{\delta R}{R}\right)^2,$$

where $A_{C_6} = 7.5\times10^{-2}$, $A_r = 3.0\times10^{-5}$, $A_x = 1.3\times10^{-3}$, and $A_R = 9.4\times10^{-2}$. Combining estimated uncertainties we obtain an error bound of 0.1% for matrix elements. This accuracy is similar to that of the high-precision direct lifetime measurements [12,13] (see Fig. 2). One notes a large discrepancy between results of the previous determinations for $\langle 6P_{1/2}||D||6S_{1/2}\rangle$; unfortunately, this discrepancy limits the conclusiveness of previous studies. Our values support experimental results by Young *et al.* [12]. The reader is referred to Ref. [13] for an extensive comparison with other measurements and theoretical predictions. Our result is most sensitive to the errors in the van der Waals coefficient. Provided that C_6 is known exactly, the uncertainty in the matrix elements may be reduced to 0.03% with our parameters.

III. CONCLUSION

We exploited a strong dependence of the van der Waals coefficient C_6 on matrix elements of principal transitions. We deduced these matrix elements by calculating small residual contributions using *ab initio* methods. Such an approach is applicable, for example, for all alkali-metal and alkaline-earth atoms. The proposed method was applied to Cs atom and the derived matrix elements have an uncertainty comparable to the best direct lifetime measurements [12,13]. Our high-precision indirect determination results in lifetimes in close agreement with the measurements [12], providing an independent reference for the required refined calculations of parity violation in Cs.

In anticipation of high-precision measurements of static dipole polarizability $\alpha(0)$ of the ground state of Cs with atom interferometry [23], we note that 96% of the polarizability is due to the contribution of the principal transitions $\alpha_p(0)$. Subtracting the residual contribution $\alpha_r^{\mathrm{se}}(0)=16.74(11)$ from exactly measured $\alpha(0)$ one may also determine matrix elements of principal transitions to an accuracy of 0.014% from Eq. (3). Alternatively, we combine contribution of principal transitions $\alpha_p(0)=383.75(80)$ computed using the derived matrix element $D_{1/2}$ and the ratio R with the $\alpha_r^{\mathrm{se}}(0)$ and obtain the ground-state static polarizability $\alpha(0)=400.49(81)$. The resulting uncertainty of 0.2% is more than a factor of 2 improvement in accuracy compared to previous determination [18], 399(1.9), and a factor of 10 improvement over measured value [24] of 402(8).

We employed the 0.36% accurate value of the van der Waals coefficient C_6 deduced by Leo *et al.* [16] from high-resolution Feshbach spectroscopy of ultracold Cs atoms [4]. According to Ref. [4] the precision of this dispersion coefficient may be potentially improved to 0.03%. If such a precision is achieved, the method proposed here, augmented with more accurate *ab initio* calculations and better measurements of the ratio R, may lead to determination of matrix elements of principal transitions with much improved 0.01% accuracy.

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