Dispersion coefficients for the interaction of Cs atom with different material media

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Largely motivated by a number of applications, the dispersion ($C_{3}$) coefficients for the interaction of a Cs atom with different material media such as Au (metal), Si (semiconductor) and various dielectric surfaces like SiO$_2$, SiNx, sapphire and YAG are determined using accurate values of the dynamic polarizabilities of the Cs atom obtained employing the relativistic coupled-cluster approach and the dynamic dielectric constants of the walls. Moreover, we also give the retardation coefficients in the graphical representation as functions of separation distances to describe the interaction potentials between the Cs atom with the above considered material media. For the easy access to the interaction potentials at a given distance of separation, we devise a simple working functional fitting form for the retarded coefficients in terms of two parameters that are quoted for each medium.

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I. INTRODUCTION

Atom-surface interactions are important for understanding numerous physical, chemical and biological processes. Owing to this, research works of Lennard-Jones \textsuperscript{1}, Bardeen \textsuperscript{2}, Casimir and Polder \textsuperscript{3} and Lifshitz \textsuperscript{4} have drawn a lot of attention over the last few decades \textsuperscript{3, 4}. In the non-retarded regime (at small distances), the forces between the fluctuating atomic dipole and its immediate image associated with the polarization charges induced in the surface brings about the atom-surface van der Waals interactions \textsuperscript{1, 10}. Thus, for the short separation distance ‘a’ between the atom and surface, the interaction energy scales as 1/a\textsuperscript{3} (at the intermediate distances, the retardation effects are taken into account by introducing a damping function), while for large atom-surface distances as compared to a typical atomic wavelength, the interaction energy scales as 1/a\textsuperscript{4} \textsuperscript{3, 11, 14}.

Investigations of the van der Waals dispersion forces between an atom and a surface are of immense interest to the physicists working in various domains of physical sciences. Assessing these forces accurately can result in new pathways towards engineering, technology and research. Finding out well behaved interactions of atoms or molecules with atomically well defined surfaces are beneficial for advocating future device applications at the nanometer dimensions \textsuperscript{12}. These atom-surface interactions can cause shifts in the oscillation frequencies of the trap that can change the trapping conditions. Therefore, when the trapping potentials are determined, particularly at the magic wavelengths, these effects need to be accounted for \textsuperscript{16}. A comprehensive cognizance of the dispersion coefficients is necessary for the experimental studies of photoassociation, realization of Bose-Einstein condensates (BECs), interpreting fluorescence spectroscopy, determination of scattering lengths, and analysis of Feshbach resonances \textsuperscript{17, 23}. Knowing their behaviors are significantly important in generating novel atom optical devices, also known as the atom chips \textsuperscript{24}.

The atom-wall interaction potentials have significant dependences on the dielectric properties of the materials \textsuperscript{25, 28}. Majority of the earlier theoretical works involve the interactions of the ground states of atoms with metallic or dielectric surfaces, considering both the non-retarded and retarded van der Waals forces \textsuperscript{1, 4, 11, 14}. Since precise measurement of the interaction potentials or in that sense the dispersion coefficients are extremely difficult, very few experimental investigations of the van der Waals interactions between the atoms in their ground states and solid state surfaces have been accomplished \textsuperscript{29}. However, very precise measurements of ratios of the dispersion coefficients of some of the alkali atoms with SiNx have been reported \textsuperscript{30, 31}. This gives an opportunity to test the validity of a method to reproduce the experimental results. For the theoretical determinations, the $C_{3}$ coefficients can be efficiently expressed in terms of the conducting properties of trapping media and the dynamic dipole polarizabilities of the interacting atom following the Lifshitz theory \textsuperscript{4, 32}. Available theoretical studies on the Cs atom-wall interactions are performed considering approximated values of either the dynamic polarizabilities of the Cs atom or the conducting properties of the materials \textsuperscript{26, 27, 33}. In our previous work, we have demonstrated importance of using accurate values of the atomic dynamic polarizabilities in order to compare ratios of the dispersion coefficients of the light nuclei alkali atoms with their corresponding measured values \textsuperscript{27}. It was shown that these interactions can be estimated with sufficient accuracy using the relativistic coupled-cluster (RCC) method.

In this work, we intend to calculate the van der Waals interaction potentials between the Cs atom in the ground state with the trapping materials as an ideal conductor, a normal good conductor like Au, a semiconductor like...
Si, dielectric objects like SiO$_2$, SiN$_x$, sapphire and a YAG surface by using accurate polarizability values of the Cs atom. Combining these results with our previously estimated $C_3$ coefficients for the other alkali atoms [23], we would like to present ratios of these constants with respect to the Cs atom. Unless stated otherwise, we use atomic units (a.u.) throughout the paper.

II. METHOD OF CALCULATIONS

The formula given by E. M. Lifshitz and collaborators in Moscow in 1955 can efficiently be applied to describe the van der Waals and Casimir-Polder interactions between an atom and a semispace, a material plate, or a layered structure [4, 32]. For the intermediate separations, where the atom-wall force of attraction is almost negligible, the atom-wall interactions can be computed by considering a polarizable particle interacting with a surface or a wall as a continuous medium having a frequency dependent permittivity $\epsilon(\omega)$. The working formula for the interaction potential in terms of the dielectric constants is given by [4, 23, 26, 32]

$$V(a) = \frac{-\alpha_f^3}{2\pi} \int_0^\infty d\omega \omega^3 \alpha(\omega) \times \int_1^\infty d\xi e^{-2\alpha_f \xi \omega a} H(\xi, \epsilon(\omega)),$$

where $\alpha_f$ is the fine structure constant, $\alpha(\omega)$ is the dynamic dipole polarizability of the atom, $a$ is the distance of separation between the Cs atom and a surface or a wall and $\epsilon(\omega)$ is the frequency dependent dielectric constant of the wall. The expression for $H(\xi, \epsilon(\omega))$, which is a function of the dielectric permittivity of the material wall, is given by

$$H(\xi, \epsilon(\omega)) = (1 - 2\xi^2) \frac{\xi - \epsilon\xi}{\xi + \epsilon\xi} + \frac{\xi^\prime - \xi}{\xi^\prime + \xi},$$

where $\xi^\prime = \sqrt{\xi^2 + \epsilon - 1}$. Evaluation procedure of this functional form is described in Refs. [23, 26]. A more general expression for the potential as described in Eq. (1) for both the retarded and short distances is conveniently expressed by

$$V(a) = \frac{-C_3}{a^3} f_3(a),$$

where the expression for the dispersion coefficient $C_3$ is given by

$$C_3 = -\frac{1}{4\pi} \int_0^\infty d\omega \alpha(\omega) S(\omega),$$

for

$$S(\omega) = \frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 1}.$$
At the short distance, this interaction potential can be approximated to a simple form as \[ V(a) = -\frac{C_3}{a^3}. \] (6)

For a perfect conductor, \( \epsilon(\omega) \rightarrow \infty \). Thus, it yields

\[ C_3 = -\frac{1}{4\pi} \int_{0}^{\infty} d\omega \alpha(\omega) \] (7)

and

\[ f_3(a) = \frac{1}{4\pi C_3} \int_{0}^{\infty} d\omega \alpha(\omega) e^{-2a_f x} P^{(\infty)}(\alpha_f x/a) \] (8)

with \( P^{(\infty)}(x) = 1 + 2x + 2x^2 \).

Adopting a similar approach as was done in Ref. [23], we actuate to evaluate the \( C_3 \) coefficients and \( f_3(a) \). We calculate the \( C_3 \) coefficients using Eq. (4) and interaction potentials using Eq. (1). By combining these two quantities, we determine the retarded function \( f_3(a) \) using Eq. (3). To calculate the \( C_3 \) and retarded functions for a perfect conductor, we use Eqs. (7) and (8) directly. As can be noticed, accurate evaluation of these quantities entirely depend on the accuracies in the values of the dynamic polarizabilities of the atom and dynamic dielectric constants of the material or a wall. Thus, importance of the present work lies in the rigorous determination of these dynamic properties of the Cs atom and materials under consideration. We describe below the approach considered to evaluate the dynamic polarizabilities of the Cs atom and dynamic dielectric constants of Au, Si, and SiO\(_2\), SiN\(_x\), sapphire and YAG surfaces having conducting, semiconducting and dielectric characteristics, respectively.

The procedure for determining accurate values of the dynamic polarizability of an atomic system having a closed core and a valence electron is given by us in Ref. [39, 40]. We apply the same procedure here to calculate the dynamic polarizabilities of the ground state of Cs. In this approach, we divide contributions to polarizability \( \alpha \) into three parts as \[ \alpha = \alpha_v + \alpha_c + \alpha_{cv} \]

where \( \alpha_v \), \( \alpha_c \) and \( \alpha_{cv} \) correspond to the polarizabilities due to the correlations due to the valence electron, core electrons and core-valence electrons correlations to the polarizability, respectively. It is known that major contributions to the alkali atoms come from \( \alpha_v \) [23, 34, 41]. We evaluate this contribution by considering predominantly contributing electric dipole (E1) matrix elements between the ground state and many low-lying excited states of Cs in a sum-over-states approach combining with the experimental energies. We use the precisely values of the E1 matrix elements for the predominantly contributing low-lying transitions estimating from the precisely measured lifetimes of the \( 6p^2 2P_{1/2} \) and \( 6p^2 2P_{3/2} \) states [33].

The other important E1 matrix elements are obtained using the RCC method in the singles and doubles approximation (CCSD method) as described in [45, 46]. Contributions to \( \alpha_v \) are determined using a relativistic random-phase approximation (RRPA) as described in [46]. It has been demonstrated that the RRPA method can give rise to very reliable results for the atomic systems having inert gas configurations [47]. Smaller contributions from

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**TABLE I**: The E1 matrix elements and various contributions to the scalar polarizability of the ground state in Cs atom.

| Transition   | E1 amplitude (a.u.) | \( \alpha \) (a.u.) |
|--------------|---------------------|---------------------|
| \( 6s_{1/2} \) - \( 6p_{1/2} \) | 4.489 [35] | 131.88 |
| \( 6s_{1/2} \) - \( 7p_{1/2} \) | 0.32 | 0.34 |
| \( 6s_{1/2} \) - \( 8p_{1/2} \) | 0.10 | 0.03 |
| \( 6s_{1/2} \) - \( 9p_{1/2} \) | 0.05 | 0.01 |
| \( 6s_{1/2} \) - \( 10p_{1/2} \) | 0.03 | \( \sim 0 \) |
| \( 6s_{1/2} \) - \( 6p_{3/2} \) | 6.324 [35] | 249.38 |
| \( 6s_{1/2} \) - \( 7p_{3/2} \) | 0.64 | 1.37 |
| \( 6s_{1/2} \) - \( 8p_{3/2} \) | 0.25 | 0.18 |
| \( 6s_{1/2} \) - \( 9p_{3/2} \) | 0.14 | 0.05 |
| \( 6s_{1/2} \) - \( 10p_{3/2} \) | 0.09 | 0.02 |
| \( \alpha_v \) | 383.30 |
| \( \alpha_c \) | 16.8 |
| \( \alpha_{cv} \) | -0.5 |
| \( \alpha_t \) | 0.2 |
| Total | 399.8 |

**TABLE II**: Calculated \( C_3 \) coefficients for the interaction of the Cs atom with the perfect conductor, Au (metal), Si (semiconductor) and the dielectric surfaces (SiO\(_2\), SiN\(_x\), Sapphire and YAG) along with the classification of contributions from various parts of the dynamic polarizabilities.

| Core | Valence | Core-Valence | Tail | Total |
|------|---------|--------------|------|-------|
| Ideal conductor | 2.350 | 2.5309 | -0.043 | 0.004 | 4.8427 |
| | | | | 4.268 [37] |
| Metal: Au | 0.706 | 2.191 | -0.017 | 0.003 | 2.8823 |
| | | | | 2.79 [26] |
| Semiconductor: Si | 0.512 | 1.874 | -0.0131 | 0.0025 | 2.3756 |
| Dielectric: | | | | |
| SiO\(_2\) | 0.310 | 0.881 | -0.0077 | 0.0012 | 1.1846 |
| SiN\(_x\) | 0.383 | 1.335 | -0.0098 | 0.0018 | 1.7100 |
| Ordinary sapphire | 0.527 | 1.319 | -0.0127 | 0.0019 | 1.8360 |
| Extraordinary sapphire | 0.551 | 1.315 | -0.0132 | 0.0019 | 1.8542 |
| Birefringent sapphire | 0.5391 | 1.317 | -0.0129 | 0.0019 | 1.84523 |
| YAG | 0.490 | 1.283 | -0.01975 | 0.0018 | 1.7635 |

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\( \alpha_{ev} \) and from the high-lying excited states (tail contribution \( \alpha_t \)) that are omitted in the above sum-over-states approach are estimated in the Dirac-Hartree-Fock (DHF) approximation.

It is not easy to get the dynamic electric permittivity of any real materials, so the convenient way of determining these constants for simple metals such as gold is to use the Drude-Lorentz model as was done in Ref. [20]. In pursuance of obtaining more realistic values of these constants for different materials, we prefer to use the known real and imaginary parts of the refractive indices of a material at few real values of frequency \( \omega \). The imaginary parts of the dielectric permittivities of the materials can then be obtained using the relation

\[
\text{Im} [\epsilon(\omega)] = 2n(\omega)\kappa(\omega),
\]

where \( n(\omega) \) and \( \kappa(\omega) \) are the respective real and imaginary parts of the refractive index of a material at frequency \( \omega \). We use the optical data from the handbook by Palik [48] for the frequencies ranging from 0.1 eV to 10000 eV for Au metal to calculate \( \text{Im} [\epsilon(\omega)] \). Thereafter, the required real values of the dielectric constants at the imaginary frequencies \( \epsilon(\omega) \) are obtained by using the Kramers-Kronig formula. The available data, however, does not cover the whole frequency range to carry out the integration of Eq. (4). Thus, we extrapolate these values for the lower frequencies to increase the domain over which the integrations are to be performed [49, 50].

Again, the interactions between the ground states of \( \text{Cs} \) atom along with the E1 matrix elements for different transitions that are used to estimate \( \alpha_v \) and other contributions to the polarizabilities. The E1 matrix elements for the \( 6S_{1/2}-6P_{1/2,3/2} \) transitions are taken from the experimentally measured data given in [42].

\[
\alpha_{ev} = 399.8 \text{ a.u.}
\]

TABLE III: Comparison of the calculated values of damping function \( f_3(a) \) defined in Eq. (11) for different separation distances \( a \) for the interaction of \( \text{Cs} \) atom with the gold surface.

| \( a \) (\text{a.u.)} | This work | Ref. [26] |
|---------------|----------|-----------|
| \( 1 \times 10^1 \) | 1.00     | 0.99780   |
| \( 2 \times 10^1 \) | 1.00     | 0.99408   |
| \( 5 \times 10^1 \) | 0.98     | 0.97903   |
| \( 1 \times 10^2 \) | 0.95     | 0.95169   |
| \( 2 \times 10^2 \) | 0.89     | 0.90210   |
| \( 5 \times 10^2 \) | 0.78     | 0.79521   |
| \( 1 \times 10^3 \) | 0.66     | 0.68309   |
| \( 2 \times 10^3 \) | 0.53     | 0.54485   |
| \( 5 \times 10^3 \) | 0.33     | 0.33918   |

where \( \epsilon || \) and \( \epsilon \bot \) are the respective dielectric permittivities for the electric fields parallel and perpendicular to the interface between the atom and the dielectric surface.

### III. RESULTS AND DISCUSSION

The evaluation of \( C_3 \) coefficients requires the precise estimation of dynamic polarizabilities of the \( \text{Cs} \) atom. Table I presents the scalar polarizabilities of the \( \text{Cs} \) atom in its ground state along with the E1 matrix elements for different transitions that are used to estimate \( \alpha_v \) and other contributions to the polarizabilities. The E1 matrix elements for the \( 6S_{1/2}-6P_{1/2,3/2} \) transitions are taken from the experimentally measured data given in [42].

Our calculated value of the scalar polarizability \( (\alpha(0)) \) for the 6s state is 399.8 a.u. This is in very good agreement with the value (399 a.u.) obtained by Borschevsky et al. [36] and experimentally measured value 401.0(6) a.u. of Amini et al. [38]. As seen, contributions from \( \alpha_{ev} \) and \( \alpha_t \) are quite small and justify use of a lower method for their evaluation. Our \( \alpha_c \) from RRPA also match with the value of Derevianko et al. [42]. Good agreement between the calculated and experimental results of \( \alpha(0) \) indicates that our approach can also give similar accuracies for the estimated dynamic polarizabilities. Next, we compute the atom-surface dispersion \( C_3 \) coefficients for a perfect conductor by numerically evaluating Eq. (4).
used an exponential grid in our calculations as discussed in Ref. [23]. In the same table we have given our calculated $C_3$ coefficients for the other material media using their dynamic dielectric constants that are plotted in Fig. IIb. In our earlier work [23], we have also given these constants for Au, Si, SiO$_2$ and SiN$_x$. Hence, we only present the final results without giving the fine details of their evaluations. Compared to these materials we find a different trend of $\epsilon(\omega)$ values for the ordinary, extraordinary and birefringent sapphires and also for the YAG surfaces, especially at the low frequency range. We could not find another study to verify directly the validity of this trend, but a recent calculation of $S(\omega)$ shows almost a similar trend. This somewhat assures us about accurate determination of the $\epsilon(\omega)$ values for the sapphires and YAG surfaces.

We also give the $C_3$ coefficients for all the considered material media in Table II along with the values known in the literature. As seen, there is another evaluation of $C_3$ for the Au metal reported by Lach et al. [26]. They use the SOM model to estimate the dynamic polarizability values whereas the dynamic dielectric constants are estimated using the Drude model. Nevertheless, we find a reasonable agreement between these results. We, however, did not find any data to compare our results for the dielectric materials and semiconductors. From Table II it can be seen that the valence correlation is dominant among the core, core-valence and tail correlations, but core contributions are quite significant as compared to their contributions in the evaluation of the polarizabilities. Among all the interacting media, the $C_3$ coefficients for the interacting perfect conductor is the highest and it is approximately 40%, 51%, 75%, 64%, 62% and 63% larger than the Au, Si, SiO$_2$, SiN$_x$, sapphire and YAG surfaces respectively. The decrease in the interaction coefficients for the cases of dielectric media might be due to the charge dangling bonds in the materials which accounts for the additional interactions in the dielectrics at the shorter separations.

Dynamic polarizabilities used for this purpose are plotted in Fig. I(a). In the same plot, we also show the values obtained by the single oscillator model (SOM) that are used by other works as discussed below. We compare our $C_3$ coefficient for the perfect conductor with the result obtained by Derevianko and co-workers [42] in Table II. Though our $\alpha(0)$ value match quite well with Derevianko et al. [37], but we find difference in the $C_3$ value. A similar finding was also observed for the other alkali atoms that were studied by us in Ref. [23]. Our analysis shows that the main reason for the discrepancy is because of different numerical integration methods used in both the works. Derevianko et al. use a Gaussian quadrature integration method with 50 point formula while we have used an exponential grid in our calculations as discussed
for the surfaces of SiO$_2$ and SiN$_x$ only as there is not very much difference in the $C_3$ coefficient values of these surfaces with those of sapphire (for all ordinary, extraordinary and birefringent) and YAG.

In Fig. 2 we show a comparison of the $f_3(a)$ values calculated for the Cs atom as a function of the atom-wall separation distance $a$ considering three material mediums such as a perfect conductor, Au (metal) and a Si (semiconductor). The retardation effects associated with the Au and Si surfaces are nearly identical at the short and intermediate separations while shows slight deviation at large distances of separation. As expected, the $f_3$ coefficients for the perfect conductor are found to be the smallest and are approximately 31% less than those of Au and Si at around 1200 a.u.. Similar plot is drawn in Fig. 3 to show the interaction potential of the Cs atom with di- and Si at around 1200 a.u.. Similar plot is drawn in Fig. 3 to show the interaction potential of the Cs atom with di-

| Surface               | $B_1$   | $B_2$   |
|-----------------------|---------|---------|
| Perfect Conductor     | 1.3077  | 0.1011  |
| Metal: Au             | 1.0563  | 0.0569  |
| Semiconductor: Si     | 1.0013  | 0.0636  |
| Dielectric: SiO$_2$   | 1.0453  | 0.0714  |
| SiN$_x$               | 0.9977  | 0.0699  |
| Ordinary Sapphire     | 1.0663  | 0.0781  |
| Extraordinary Sapphire| 1.0741  | 0.0809  |
| Birefringent Sapphire | 1.0702  | 0.0777  |
| YAG                   | 1.0535  | 0.0776  |

for the above interactions and then studied the retardation effects of the above interactions by plotting the damping coefficient $f_3$ values as function of separation distances. More accurate dynamic polarizabilities of Cs atom are used in determining these coefficients. Furthermore, readily usable functional form of the retardation coefficients for the above atom-surface interactions is devised and the fitting parameters are given.

IV. CONCLUSION

In the present work, we have analyzed the atom-surface interactions between the cesium atom in its ground state with a perfect conductor, Au (metal), Si (semiconductor) and various dielectric surfaces (SiO$_2$, SiN$_x$, sapphire and YAG). We have calculated the dispersion $C_3$ coefficients for the above interactions and then studied the retardation effects of the above interactions by plotting the damping coefficient $f_3$ values as function of separation distances. More accurate dynamic polarizabilities of Cs atom are used in determining these coefficients. Furthermore, readily usable functional form of the retardation coefficients for the above atom-surface interactions is devised and the fitting parameters are given.

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