Polarizabilities of Si\(^{2+}\): A benchmark test of theory and experiment

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Polarizabilities of Si$^{2+}$: a benchmark test of theory and experiment

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We have calculated electric-dipole polarizabilities of the 3$s^2$ $^1S_0$, 3$s 3p$ $^3P_0$, and 3$s 3p$ $^1P_1$ states of the Si$^{2+}$ ion using recently developed configuration interaction + all-order method. Detailed evaluation of the uncertainties of the final results is carried out. Our value for the ground state electric-dipole polarizability 11.670(13) a.u. is in excellent agreement with the resonant excitation Stark ionization spectroscopy value 11.669(9) a.u. [Komara et al., J. Phys. B 38, 87 (2005); Mitroy, Phys. Rev. A 78, 052515 (2008)]. This work represents the most precise benchmark test to date of theory and experiment in divalent atoms. The near cancellation of the $n$s$n$p $^3P_0$ polarizabilities previously observed in B$^+$, Al$^+$, In$^+$, Ti$^+$, and Pb$^{2+}$ is also found in Si$^{2+}$ ion.

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I. INTRODUCTION

The atomic dipole polarizability describes the first-order response of an atom to an applied electric field. Atomic polarizabilities have been the subject of considerable interest and heightened importance in recent years due to a number of applications, including development of next-generation optical atomic clocks, optical cooling and trapping schemes, quantum information with atoms and ions, tests of fundamental symmetries, studies of cold degenerate gases, thermometry and other macroscopic order response of an atom to an applied electric field. Atomic polarizabilities have been the subject of considerable interest and heightened importance in recent years due to a number of applications, including development of next-generation optical atomic clocks, optical cooling and trapping schemes, quantum information with atoms and ions, tests of fundamental symmetries, studies of cold degenerate gases, thermometry and other macroscopic

A. Experimental determination of polarizabilities from Rydberg spectra

The polarizability of an ion can be extracted from the energies of the non-penetrating Rydberg series of the corresponding parent system (see [19] and references therein). The polarization interaction between the ionic core and the Rydberg electron shifts the energy levels away from their hydrogenic values. If the Rydberg electron is in a high angular momentum state, it has negligible overlap with the core. In such cases, the polarization interaction provides the dominant contribution to the energy shift. This effect is utilized in resonant excitation Stark ionization spectroscopy (RESIS) [19–28]. RESIS experiments have been extremely successful in high-precision determination of the ground state polarizabilities of H$_2^+$ and D$_2^+$ [21], Ne$^+$ [19], Na-like Mg$^+$ [22], Na-like Si$^{3+}$ [23], Mg-like Si$^{2+}$ [20], Zn-like K$^{6+}$ [28], Ba$^+$ [24], Hg-like Pb$^{2+}$ [25], Fr-like Th$^{3+}$ [26], and Ru-like Th$^{3+}$ [27]. Quadrupole polarizabilities and transition matrix elements have also been determined for some of these systems.

To the best of our knowledge, RESIS experiments provide the most precise values known to date of the polarizability of any divalent atomic system. The most precise measurement has been carried out for the 3$s^2$ $^1S_0$ ground state of the Si$^{2+}$ ion; $\alpha_0 = 11.666(4)$ a.u [20]. Later analysis of the RESIS data that included additional terms in the polarization expansion yielded $\alpha_0 = 11.669(9)$ a.u [29]. Therefore, the Si$^{2+}$ RESIS experiment presents an excellent opportunity for a high-precision benchmark comparison of theory and experiment.

In this work, we use a recently developed configuration iteration (CI)+ all-order method [30–32] to calculate properties of Si$^{2+}$. Our value for the ground state electric-dipole polarizability of 11.670(13) a.u. is in excellent agreement with the RESIS result. Our previous calculation of the Hg-like Pb$^{2+}$ ground state polarizabil-
ity [33] was also in agreement with the RESIS value (accurate to 0.6%) within our estimated accuracy.

We note that Mg-like Si$^{2+}$ is a particularly interesting test system due to its similarity with Mg-like Al$^+$, which was used to construct an optical clock with a fractional frequency uncertainty of $8.6 \times 10^{-18}$ [35], the smallest such uncertainty yet attained. At room temperature, one of the largest contributions to the uncertainty budget of this clock is the blackbody radiation (BBR) shift. The BBR frequency shift of a clock transition is related to the difference of the static electric-dipole polarizabilities between the two clock states [36]. We have recently calculated this effect in Al$^+$ using the same CI+all-order approach. Excellent agreement of our present calculation with the experiment in the ground state of Si$^{2+}$ provides an additional test of the approach. At the present time, there are no experimental data on the polarizabilities of the excited states of Si$^{2+}$ to the best of our knowledge.

II. METHOD

To evaluate uncertainties of the final results, we carry out three calculations in different approximations: CI [37], CI+many-body perturbation theory (MBPT) [38], and CI+all-order [30–32]. These methods have been described in a number of papers [30, 31, 37, 38] and we provide only a brief outline of these approaches and a few details relevant to this particular work.

Our point of departure is a solution of the Dirac-Fock (DF) equations

$$
\hat{H}_0 \psi_c = \varepsilon_c \psi_c,
$$

where $H_0$ is the relativistic DF Hamiltonian [31, 38] and $\psi_c$ and $\varepsilon_c$ are single-electron wave functions and energies. Self-consistent calculations were performed for the $[1s^22s^22p^6]$ closed core, and the 3s, 3p, 3d, 4s, 4p, and 4d orbitals were formed in this potential. We constructed the B-spline basis set consisting of $N = 35$ orbitals for each of the s, p$_{1/2}$, p$_{3/2}$, ... partial waves up to $l \leq 5$. The basis set is formed in a spherical cavity with radius 60 a.u. The CI space is effectively complete and includes 23 orbitals for each partial wave with $l = 0 \ldots 4$.

The wave functions and the low-lying energy levels are determined by solving the multiparticle relativistic equation for two valence electrons [37]:

$$
H_{\text{eff}}(E_n) \Phi_n = E_n \Phi_n.
$$

The effective Hamiltonian is defined as

$$
H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E),
$$

where $H_{\text{FC}}$ is the Hamiltonian in the frozen-core approximation. The energy-dependent operator $\Sigma(E)$ takes into account virtual core excitations. It is zero in a pure CI calculation. The $\Sigma(E)$ part of the effective Hamiltonian is constructed using second-order perturbation theory in the CI+MBPT approach [38] and linearized coupled-cluster single-double method in the CI+all-order approach [31]. Construction of the effective Hamiltonian in the CI+MBPT and CI+all-order approximations is described in detail in Refs. [31, 38]. The dominant part of the Breit interaction is included as described in Ref. [39].

The scalar polarizability $\alpha_0$ is separated into a valence polarizability $\alpha_v^{\text{eff}}$, ionic core polarizability $\alpha_c$, and a small term $\alpha_{vc}$ that modifies ionic core polarizability due to the presence of two valence electrons. The last two terms are evaluated in the random-phase approximation (RPA). Their uncertainty is determined by comparing the DF and RPA values. The small $\alpha_{vc}$ term is calculated by adding vc contributions from the individual electrons, i.e.

$$
\alpha_{vc}(3s^2) = 2 \alpha_{vc}(3s), \quad \text{and} \quad \alpha_{vc}(3s3p) = \alpha_{vc}(3s) + \alpha_{vc}(3p).
$$

The valence part of the polarizability is determined by solving the inhomogeneous equation in valence space, which is approximated as [40]

$$
(E_n - H_{\text{eff}}) | \Psi(v, M') \rangle = D_{\text{eff}} | \Psi_0(v, J, M) \rangle
$$

for the state $v$ with total angular momentum $J$ and projection $M$. The wave function $\Psi(v, M')$ is composed of parts that have angular momenta of $J' = J \pm 1$ that allows us to determine the scalar and tensor polarizability of the state $|v, J, M \rangle$ [40]. The effective dipole operator $D_{\text{eff}}$ includes RPA corrections.

Unless stated otherwise, we use atomic units (a.u.) for all matrix elements and polarizabilities throughout this paper: the numerical values of the elementary charge, e, the reduced Planck constant, $\hbar = \hbar/2\pi$, and the electron mass, $m_e$, are set equal to 1. The atomic unit for polarizability can be converted to SI units via $\alpha/\hbar \left[ \text{Hz}/(\text{V/m})^2 \right] = 2.48832 \times 10^{-6} \alpha$ (a.u.), where the conversion coefficient is $4\pi\varepsilon_0 a_0^3/\hbar$ and the Planck constant $\hbar$ is factored out in order to provide direct conversion into frequency units; $a_0$ is the Bohr radius and $\varepsilon_0$ is the electric constant.

III. RESULTS

Comparison of the energy levels (in cm$^{-1}$) obtained in the CI, CI+MBPT, and CI+all-order approximations with experimental values [34] is given in Table I. Corresponding relative differences of these three calculations from experiment are given in the last three columns. Two-electron binding energies are given in the first row of Table I, energies in other rows are measured from the ground state. For a few of the levels, the accuracy of the CI+MBPT calculation is already on the order of our expected precision. The accuracy of the ground state two-electron binding energy is significantly improved in the CI+all-order calculation in comparison with the CI+MBPT one; the CI+MBPT value differs from the experiment by $-123$ cm$^{-1}$, while our all-order value differs from the experiment by only $-7$ cm$^{-1}$ (see line one of Table I). The inclusion of the all-order correlation coefficients significantly improves the differences
TABLE I: Comparison of experimental [34] and theoretical energy levels in cm$^{-1}$. Two-electron binding energies are given in the first row, energies in other rows are given relative to the ground state. Results of the CI, CI+MBPT, and CI+all-order calculations are given in columns labeled CI, CI+MBPT, and CI+All. Corresponding relative differences of these three calculations with experiment are given in the last three columns in %.

| State      | Expt. | CI   | CI+MBPT | CI+All | Differences (cm$^{-1}$) | Differences (%) |
|------------|-------|------|---------|--------|-------------------------|-----------------|
| 3$s^2$ 1$S_0$ | 634232 | 628511 | 634110 | 634226 | -5722 | -123 | -7 | -0.9% | -0.019% | -0.001% |
| 3p$^2$ 1$D_2$ | 122215 | 120224 | 122225 | 122294 | -1901 | 10 | 80 | -1.6% | 0.088% | 0.065% |
| 3p$^2$ 3$P_0$ | 129708 | 128589 | 129745 | 129753 | -1119 | 36 | 45 | -0.9% | 0.028% | 0.035% |
| 3p$^2$ 3$P_1$ | 129842 | 128717 | 129878 | 129887 | -1125 | 36 | 45 | -0.9% | 0.028% | 0.035% |
| 3p$^2$ 3$P_2$ | 130101 | 128964 | 130136 | 130145 | -1137 | 35 | 44 | -0.9% | 0.027% | 0.034% |
| 3$s^2$ 3$D_3$ | 142944 | 141676 | 142953 | 142944 | -1268 | 10 | 1 | -0.9% | 0.007% | 0.000% |
| 3$s^2$ 3$D_2$ | 142946 | 141678 | 142955 | 142946 | -1267 | 10 | 1 | -0.9% | 0.007% | 0.000% |
| 3$s^2$ 3$D_1$ | 142948 | 141681 | 142957 | 142948 | -1268 | 9 | 0 | -0.9% | 0.006% | 0.000% |
| 3$s^2$ 3$S_1$ | 153377 | 151756 | 153357 | 153403 | -1621 | 20 | 26 | -1.1% | 0.013% | 0.017% |
| 3p$^2$ 1$S_0$ | 153444 | 152674 | 153631 | 153613 | -771 | 187 | 169 | -0.5% | 0.122% | 0.110% |
| 3$s^2$ 1$S_0$ | 159070 | 157543 | 159079 | 159116 | -1527 | 9 | 47 | -1.0% | 0.006% | 0.029% |
| 3$s^2$ 1$D_2$ | 165765 | 165071 | 165937 | 165898 | -694 | 172 | 133 | -0.4% | 0.104% | 0.080% |
| 3$s^2$ 3$D_3$ | 52725 | 51559 | 52722 | 52770 | -1166 | -3 | 3 | -2.2% | -0.006% | 0.086% |
| 3$s^2$ 3$P_0$ | 52853 | 51682 | 52849 | 52897 | -1171 | -4 | 4 | -2.2% | -0.008% | 0.083% |
| 3$s^2$ 3$P_1$ | 53115 | 51934 | 53110 | 53159 | -1181 | -5 | 4 | -2.2% | -0.010% | 0.082% |
| 3$s^2$ 3$P_2$ | 82884 | 82998 | 82969 | 82933 | 113 | 84 | 48 | 0.1% | 0.102% | 0.058% |
| 3$s^2$ 3$D_0$ | 175230 | 173409 | 175202 | 175249 | -1821 | -28 | 19 | -1.0% | -0.016% | 0.011% |
| 3$s^2$ 3$D_1$ | 175263 | 173441 | 175235 | 175282 | -1822 | -28 | 19 | -1.0% | -0.016% | 0.011% |
| 3$s^2$ 3$P_2$ | 175336 | 173511 | 175308 | 175355 | -1825 | -28 | 18 | -1.0% | -0.016% | 0.011% |
| 3$s^2$ 3$D_1$ | 176487 | 174807 | 176469 | 176511 | -1680 | -18 | 23 | -1.0% | -0.010% | 0.013% |

between the singlet and triplet states. For example, the CI+all-order value of the 3$s^2$ 1$S_0$, 3$s^2$ 3$D_3$, and 3$s^2$ 3$P_0$ energy splittings are 136 cm$^{-1}$ and 413 cm$^{-1}$, respectively. The differences between the CI+all-order and experimental values are given in the last three columns in %.

We separated the effect of the Breit interaction by comparing the results of the calculations with and without the Breit. The Breit contribution to the energies is very small, 0.01% or less. However, the inclusion of the Breit interaction significantly improves the splittings of all triplet states. For example, the 3$s^2$ 3$P_1$-3$s^2$ 3$P_0$ and 3$s^2$ 3$P_2$-3$s^2$ 3$P_1$ splittings are 136 cm$^{-1}$ and 413 cm$^{-1}$ without Breit, respectively. The values of these splittings in our final calculations include Breit, are 128 cm$^{-1}$ and 389 cm$^{-1}$, in excellent agreement with the experimental values, 129 cm$^{-1}$ and 390 cm$^{-1}$.

We note that the transition energies relevant to the calculations of the 3$s^2$ 3$P_0$ polarizabilities are more accurate than the energies relative to the ground state listed in Table I.

While we do not use the sum-over-state approach in the calculation of the polarizabilities, it is useful to establish the dominant contributions to the final values. We combine our CI+all-order results for the electric-dipole matrix elements and energies according to the sum-over-states formula for the valence polarizability [1]:

$$\alpha_v(A) = \frac{2}{3(2J+1)} \sum_n \frac{|\langle v|D|n\rangle|^2}{E_n - E_v}$$

(2)
to calculate the contribution of specific transitions. Here, $J$ is the total angular momentum of the state $v$, $D$ is the electric-dipole operator, and $E_i$ is the energy of the state $i$. The breakdown of the contributions to the 3$s^2$ 1$S_0$, 3$s^2$ 3$D_0$, and 3$s^2$ 3$P_0$ scalar polarizabilities $\alpha_v$ of Si$^{2+}$ in a.u. is given in Table II. Absolute values of the corresponding reduced electric-dipole matrix elements are listed in column labeled “$D$” in $\alpha_v$. The theoretical and experimental [34] transition energies are given in columns $\Delta E_{th}$ and $\Delta E_{expt}$. The remaining valence contributions are given in rows. The contributions from the core and $v$-terms are listed in rows with “ cores”, respectively.

The dominant contributions to $\alpha_v$ listed in columns $\alpha_v(A)$ and $\alpha_v(B)$ are calculated with CI + all-order energies and experimental [34] energies, respectively. The differences between $\alpha_v(A)$ and $\alpha_v(B)$ values are small due to excellent agreement of the corresponding transition energies with experiment. We take $\alpha_v(B)$ results as final. Our study of the Breit interaction shows that it contributes only 0.03-0.07% to the $ab$ initio values of polarizabilities.
The dominant contributions to the valence polarizabilities are listed separately with the corresponding absolute values of electric-dipole reduced matrix elements given in columns labeled $D$. The theoretical and experimental [34] transition energies are given in columns $\Delta E_{\text{th}}$ and $\Delta E_{\text{expt}}$. The remaining contributions to valence polarizability are given in rows Other. The contributions from the core and $\text{vc}$ terms are given in rows $\alpha_0$ and $\alpha_{\text{vc}}$, respectively. The dominant contributions to $\alpha_0$ listed in columns $\alpha_0(A)$ and $\alpha_0(B)$ are calculated with CI + all-order and experimental energies [34], respectively.

**TABLE II:** Contributions to the $3s^2 1S_0$, $3s3p^3 P_0$, and $3s3p^1 P_1$ polarizabilities of Si$^{2+}$ in a.u. The dominant contributions to the valence polarizabilities are listed separately with the corresponding absolute values of electric-dipole reduced matrix elements given in columns labeled $D$. The theoretical and experimental [34] transition energies are given in columns $\Delta E_{\text{th}}$ and $\Delta E_{\text{expt}}$. The remaining contributions to valence polarizability are given in rows Other. The contributions from the core and $\text{vc}$ terms are given in rows $\alpha_0$ and $\alpha_{\text{vc}}$, respectively. The dominant contributions to $\alpha_0$ listed in columns $\alpha_0(A)$ and $\alpha_0(B)$ are calculated with CI + all-order and experimental energies [34], respectively.

| State         | Contribution          | $\Delta E_{\text{expt}}$ | $\Delta E_{\text{th}}$ | $D$   | $\alpha_0(A)$ | $\alpha_0(B)$ |
|---------------|-----------------------|---------------------------|------------------------|-------|----------------|----------------|
| $3s^2 1S_0$   | $3s^2 1S_0 - 3s3p^1 P_1$ | 82884                     | 82933                  | 2.539 | 11.375         | 11.382         |
| $3s^2 1S_0$   | $3s^2 1S_0 - 3s3p^1 P_1$ | 176477                    | 176511                 | 0.198 | 0.032          | 0.032          |
| Other         |                       |                           |                        |       | 0.105          | 0.105          |
| $\alpha_e$    |                       |                           |                        |       | 0.162          | 0.162          |
| $\alpha_{\text{vc}}$ |                  |                           |                        |       | -0.011         | -0.011         |
| Total         |                       |                           |                        |       | 11.664         | 11.670         |
| $3s3p^3 P_0$  | $3s3p^3 P_0 - 3p^2 3P_1$ | 77117                     | 77117                  | 1.516 | 4.359          | 4.359          |
| $3s3p^3 P_0$  | $3s3p^3 P_0 - 3s3d 3D_1$ | 90224                     | 90179                  | 1.779 | 5.137          | 5.135          |
| $3s3p^3 P_0$  | $3s3p^3 P_0 - 3s4s 3S_1$ | 100652                    | 100633                 | 0.628 | 0.573          | 0.573          |
| Other         |                       |                           |                        |       | 0.201          | 0.201          |
| $\alpha_e$    |                       |                           |                        |       | 0.162          | 0.162          |
| $\alpha_{\text{vc}}$ |                  |                           |                        |       | -0.006         | -0.006         |
| Total         |                       |                           |                        |       | 10.427         | 10.425         |
| $3s3p^1 P_1$  | $3s3p^1 P_1 - 3s^2 1S_0$ | -82884                    | -82933                 | 2.539 | -3.792         | -3.794         |
| $3s3p^1 P_1$  | $3s3p^1 P_1 - 3p^2 1D_2$ | 39330                     | 39361                  | 1.074 | 1.428          | 1.429          |
| $3s3p^1 P_1$  | $3s3p^1 P_1 - 3s^2 1S_0$ | 70560                     | 70680                  | 1.776 | 2.178          | 2.181          |
| $3s3p^1 P_1$  | $3s3p^1 P_1 - 3s4s 1S_0$ | 76185                     | 76184                  | 0.996 | 0.634          | 0.634          |
| $3s3p^1 P_1$  | $3s3p^1 P_1 - 3s3d 1D_2$ | 82881                     | 82965                  | 4.450 | 11.642         | 11.654         |
| Other         |                       |                           |                        |       | 0.440          | 0.440          |
| $\alpha_e$    |                       |                           |                        |       | 0.162          | 0.162          |
| $\alpha_{\text{vc}}$ |                  |                           |                        |       | -0.006         | -0.006         |
| Total         |                       |                           |                        |       | 12.686         | 12.701         |

**IV. EVALUATION OF THE UNCERTAINTY AND CONCLUSION**

There are three contributions to the uncertainties in the final polarizability values that arise from the uncertainties in the valence $\alpha_0^v$, core $\alpha_e$, and $\text{vc} \alpha_{\text{vc}}$ polarizability terms. To evaluate uncertainty in the valence polarizabilities, we compare the results of the CI, CI+MBPT, CI+all-order calculations with our final CI+all-order calculation in which energies in the dominant contributions are replaced by their experimental values. The results of the last two calculations are given in Table II in columns $\alpha_0(A)$ and $\alpha_0(B)$. We summarize the results of all four calculations in Table III. For consistency, we refer to these calculations as CI (A), CI+MBPT (A), CI+All (A), and CI+All (B) since only theoretical energies (in the corresponding approximation) were used in the first three calculations. We evaluate the uncertainty of the final results in two different ways: (1) as the difference between the CI+all-order and CI+MBPT calculations, listed in row labeled Diff. (All-MBPT), and (2) as the difference between the CI+all-order results with theoretical and experimental energies, listed in row labeled Diff. (B)-(A). We take the largest of the two uncertainties as the final uncertainty in the valence polarizability $\alpha_0^v$. The uncertainty analysis is carried out separately for each state.

aThis value is a result of revised analysis [29] of the RESIS experiment [20].
To evaluate the uncertainty in the $\alpha_c$ and $\alpha_{nc}$ contribution to the polarizability, we calculate these terms in both DF and RPA approximations. The DF values for the $\alpha_c$ and $\alpha_{nc}(3s^2)$ are 0.153 a.u. and -0.0086 a.u., respectively. The difference between the RPA and DF results is taken to be the uncertainty. Uncertainties of the core and valence polarizabilities are added in quadrature to obtain uncertainties of the final values.

The final results listed in row labelled “Total $\alpha_0$” are compared with other theory [29, 41] and experiment [20, 29]. Our value for the ground state polarizability is in excellent agreement with both original RESIS value [20] value and the revised RESIS analysis [29]. Our values for the ground and $3s3p \, ^1P_1$ state polarizabilities are in excellent agreement with theoretical values obtained with large-scale CI calculation with semiempirical inclusion of the core polarization [29]. The CI result of [41] is consistent with other values; the small difference is probably due to omission of the highly-excited states in the valence CI and restricted treatment of the core excitations in [41].

We note that the values of the $^1S_0$ and $^3P_0$ polarizabilities given in Table III are very similar, their difference is only 10% of the ground state polarizability.

In summary, we have carried out a benchmark test of the theoretical and experimental determination of the ground state polarizability of the Si$^{2+}$ ion. Our final result is in excellent agreement with the RESIS experimental value [20, 29]. High-precision recommended values are provided for the excited state $3s3p \, ^3P_0$ and $3s3p \, ^1P_1$ polarizabilities. The near cancellation of the $ns^2 \, ^1S_0$ ground state and the lowest $nsnp \, ^1P_0$ polarizabilities reported for B$^+$, Al$^+$, In$^+$, Tl$^+$, and Pb$^{2+}$ is also observed for the Si$^{2+}$ ion.

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