

Multipolar Polarizabilities and Hyperpolarizabilities in the Sr Optical Lattice Clock

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We address the problem of the lattice Stark shifts in the Sr clock caused by the multipolar $M1$ and $E2$ atom-field interactions and by the term nonlinear in lattice intensity and determined by the hyperpolarizability. We develop an approach to calculate hyperpolarizabilities for atoms and ions based on a solution of the inhomogeneous equation which allows us to effectively and accurately carry out complete summations over intermediate states. We apply our method to the calculation of the hyperpolarizabilities for the clock states in Sr. We also carry out an accurate calculation of the multipolar polarizabilities for these states at the magic frequency. Understanding these Stark shifts in optical lattice clocks is crucial for further improvement of the clock accuracy.

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Introduction.—Extraordinary advancements in optical atomic clock accuracy and stability have been demonstrated in the past few years [1–6]. The systematic uncertainty of the Sr optical lattice clock was reduced to 2.1×10^{-18} in fractional frequency units [3]. A similar systematic uncertainty of 3.2×10^{-18} was reported in a single trapped ion atomic clock based on the electric-octupole transition in Yb^+ [4]. A fractional frequency instability of $6 \times 10^{-17}/\sqrt{\tau}$ for an averaging time τ in seconds was demonstrated in Ref. [5] using a zero-dead-time optical clock based on the interleaved interrogation of two cold-atom ensembles. A Fermi-degenerate three-dimensional optical lattice clock was demonstrated in Ref. [6], with a synchronous clock comparison between two regions of the 3D lattice yielding a measurement precision of 5×10^{-19} in 1 hour of averaging time. Improved precision of atomic clocks enables many applications, including relativistic geodesy [7], very long baseline interferometry [8], search for variations of the fundamental constants [9] and dark matter [10,11], tests of the Lorentz invariance [12], redefinition of the second [13], and others. Further improvement of clock precision is needed for these applications and the implementation of new ideas, such as the use of atomic clocks for gravitational wave detection [14].

When an atom is placed in a laser field, the atomic energy levels shift due to the ac Stark effect. In the main approximation, these shifts are determined by the frequency-dependent electric dipole polarizabilities of the atomic states [15]. To cancel the ac Stark shift of the clock transition, the trap laser of the optical lattice clock is operated at the “magic wavelength” [16,17], at which the electric dipole polarizabilities of the clock states are the

same, resulting in zero ac Stark shift of a clock transition to some degree of precision.

With the clock systematic uncertainties reaching 10^{-18} , such cancellation is no longer sufficient, as other contributions to the Stark shift effects become significant [18–20]: the magnetic-dipole ($M1$) and electric-quadrupole ($E2$) interactions of the atom with the lattice field and the term nonlinear in the trap laser intensity and determined by the hyperpolarizability.

Measurements and calculations of these effects are both very difficult [20], with contradictory results obtained for Sr [3,18,19,21,22] for both theory and experiment. Theoretical evaluations of the $M1$ and $E2$ polarizabilities in Sr disagree even in the sign of the effect [18,19]. The experimental determination of the Sr clock differential hyperpolarizability $\Delta\beta = \beta(^3P_0^o) - \beta(^1S_0)$ at the magic frequency by JILA [3] is consistent with zero [$\Delta\beta = -1.3(1.3) \times 10^7$ a.u.], but the SYRTE [21,22] measurement [$-2.01(45) \times 10^7$ a.u.] is not. Recent experimental assessment of the nonlinear hyperpolarizability ac Stark shifts in Yb found a difficulty associated with the finite-temperature effects [20]. In summary, reliable theoretical values of the ac multipolar polarizabilities and hyperpolarizability are required to control lattice shifts at the 10^{-19} level.

So far, there are no reliable calculations of the clock states’ hyperpolarizabilities for divalent atoms. An accurate calculation of hyperpolarizabilities is very difficult, because certain terms are expressed by complicated formulas, involving three summations over intermediate states, which cannot be properly evaluated by summing over low-lying contributions. An exception is the case of Yb, where the

magic wavelength happens to be accidentally close to relevant atomic transition energies causing strong resonant effects, and making a very few hyperpolarizability terms dominant. For Sr, however, the full expression must be evaluated, and we focus on this example.

In this Letter, we develop a method that allows us to solve this problem and accurately calculate the hyperpolarizabilities for states with the total angular momentum $J = 0$, which includes Sr, Yb, Hg, Mg, and Cd clock cases. A generalization of the method to higher total angular momenta is straightforward but technically more complicated. We apply our method to calculate the 1S_0 and $^3P_0^o$ clock states' hyperpolarizabilities at the magic frequency in Sr and their uncertainties. Our results definitively determine that the Sr hyperpolarizability is not zero and has to be taken into account when reducing lattice shifts at the next level of precision. We also calculate Sr clock magnetic-dipole and electric-quadrupole polarizabilities, resolving previous major discrepancies and providing the final differential multipolar polarizability with only 13% uncertainty.

General formalism.—We assume that an atom in a state $|0\rangle \equiv |\gamma_0, J_0 = 0\rangle$ (where γ_0 encapsulates all other quantum numbers) is placed in a linear polarized field of the lattice standing wave with the electric-field vector along the z axis, given by

$$\mathcal{E}_z = 2(\mathcal{E}_0)_z \cos(kx) \cos(\omega t). \quad (1)$$

Here $k = \omega/c$, ω is the lattice laser wave frequency, c is the speed of light, and the factor 2 accounts for the superposition of forward and backward traveling along the x -axis waves that doubles the lattice standing-wave amplitude. The atom-lattice interaction can be written as $V(x, t) = \text{Re}[V(x)e^{-i\omega t}]$, with the spatial part $V(x)$ determined by the expression [18,19]

$$V(x) = V_{E1} \cos(kx) + (V_{M1} + V_{E2}) \sin(kx). \quad (2)$$

Here V_{E1} , V_{M1} , and V_{E2} correspond to operators of $E1$, $M1$, and $E2$ interactions, and x determines the position of the atom starting from the standing-wave antinode.

The optical lattice potential for the atom at $|kx| \ll 1$ can be approximated as [18,19]

$$\begin{aligned} U(\omega) \approx & -\alpha^{E1}(\omega)(1 - k^2x^2)\mathcal{E}_0^2 \\ & - \{\alpha^{M1}(\omega) + \alpha^{E2}(\omega)\}k^2x^2\mathcal{E}_0^2 \\ & - \beta(\omega)(1 - 2k^2x^2)\mathcal{E}_0^4. \end{aligned} \quad (3)$$

The ac 2^K -pole polarizability of the $|0\rangle$ state is expressed (we use atomic units $\hbar = m = |e| = 1$) as [23]

$$\begin{aligned} \alpha^{\lambda K}(\omega) = & \frac{K+1}{K} \frac{2K+1}{[(2K+1)!!]^2} (\alpha\omega)^{2K-2} \\ & \times \sum_n \frac{(E_n - E_0) |\langle n || T_{\lambda K} || 0 \rangle|^2}{(E_n - E_0)^2 - \omega^2}. \end{aligned} \quad (4)$$

Here λ distinguishes between electric ($\lambda = E$) and magnetic ($\lambda = M$) multipoles, and $\langle n || T_{\lambda K} || 0 \rangle$ are the reduced matrix elements of the multipole operators, $T_{E1} \equiv D$, $T_{M1} \equiv \mu$, and $T_{E2} \equiv Q$.

The hyperpolarizability $\beta(\omega)$ of the $|0\rangle$ state is determined as

$$\beta(\omega) = \frac{1}{9} Y_{101}(\omega) + \frac{2}{45} Y_{121}(\omega). \quad (5)$$

The quantities $Y_{101}(\omega)$ and $Y_{121}(\omega)$ are given by

$$\begin{aligned} Y_{101}(\omega) \equiv & \sum_q \mathcal{R}_{101}(q\omega, 2q\omega, q\omega) \\ & + \sum_{q,q'} [\mathcal{R}'_{101}(q\omega, 0, q'\omega) - \mathcal{R}_1(q'\omega)\mathcal{R}_1(q\omega, q\omega)], \\ Y_{121}(\omega) \equiv & \sum_q \left(\mathcal{R}_{121}(q\omega, 2q\omega, q\omega) \right. \\ & \left. + \sum_{q'} \mathcal{R}_{121}(q\omega, 0, q'\omega) \right), \end{aligned}$$

where $q, q' = \pm 1$ and

$$\mathcal{R}_{1J_n1}(\omega_1, \omega_2, \omega_3) \equiv \sum_{\gamma_m, \gamma_n, \gamma_k} \frac{\langle 0 || D || \gamma_m, 1 \rangle \langle \gamma_m, 1 || D || \gamma_n, J_n \rangle \langle \gamma_n, J_n || D || \gamma_k, 1 \rangle \langle \gamma_k, 1 || D || 0 \rangle}{(E_m - E_0 - \omega_1)(E_n - E_0 - \omega_2)(E_k - E_0 - \omega_3)}, \quad (6)$$

$$\begin{aligned} \mathcal{R}_1(\omega) & \equiv \sum_{\gamma_m} \frac{|\langle 0 || D || \gamma_m, 1 \rangle|^2}{E_m - E_0 - \omega}, \\ \mathcal{R}_1(\omega, \omega) & \equiv \sum_{\gamma_k} \frac{|\langle 0 || D || \gamma_k, 1 \rangle|^2}{(E_k - E_0 - \omega)^2}. \end{aligned} \quad (7)$$

The notation \mathcal{R}'_{101} (i.e., the prime over \mathcal{R}) means that the term $|\gamma_n, 0\rangle = |\gamma_0, 0\rangle$ must be excluded from the summation over γ_n in Eq. (6).

The term $\mathcal{R}_{1J_n1}(\omega_1, \omega_2, \omega_3)$ contains three summations over intermediate states which so far precluded accurate calculation of the hyperpolarizabilities for lattice clocks. A large number of the intermediate states should be included in the summation unless some accidental resonances arise, making such an approach impractical.

In this Letter, we develop another method of calculating hyperpolarizabilities, which is based on solutions of the inhomogeneous equations. We consider

$$\begin{aligned}\alpha_{q\omega}(\gamma_0, 0) &= \frac{2}{3} \sum_{\gamma_m} \frac{|\langle 0 \| D \| \gamma_m, 1 \rangle|^2}{E_m - E_0 - q\omega} \\ &= \frac{2}{3} \sum_{\gamma_m} \frac{\langle 0 \| D \| \gamma_m, 1 \rangle \langle \gamma_m, 1 \| D \| 0 \rangle}{E_0 - E_m + q\omega},\end{aligned}\quad (8)$$

which can be treated as the scalar static electric dipole polarizability of the $|0\rangle$ state, calculated at the shifted energy $(E_0 + q\omega)$. For brevity, we omit the superscript $E1$, i.e., $\alpha_{q\omega} \equiv \alpha_{q\omega}^{E1}$. We use the Sternheimer [24] or Dalgarno-Lewis [25] method and solve the inhomogeneous equation

$$(E_0 - H_{\text{eff}} + q\omega)|\delta\phi\rangle = D_z|0\rangle, \quad (9)$$

where H_{eff} is the effective Hamiltonian determined below [see Eq. (13)]. Solving Eq. (9) and introducing $|\delta\phi_q\rangle$ as

$$|\delta\phi_q\rangle \equiv \sqrt{3}|\delta\phi\rangle = \sum_{\gamma_m} \frac{|\gamma_m, 1\rangle \langle \gamma_m, 1 \| D \| 0 \rangle}{E_0 - E_m + q\omega},$$

we find $\alpha_{q\omega}(\gamma_0, 0)$ as the expectation value

$$\alpha_{q\omega}(\gamma_0, 0) = \frac{2}{3} \langle 0 \| D \| \delta\phi_q \rangle. \quad (10)$$

Substituting $|\delta\phi_q\rangle$ into Eq. (6), we obtain

$$\mathcal{R}_{1J_n,1}(q\omega, 2q\omega, q\omega) = \sum_{\gamma_n} \frac{|\langle \delta\phi_q \| D \| \gamma_n, J_n \rangle|^2}{E_n - E_0 - 2q\omega}. \quad (11)$$

Thus, using this approach, we include all discrete and continuum intermediate states in the sums over γ_m and γ_k in Eq. (6). A direct summation over all intermediate states at fixed values $J_n = 0, 2$ in Eq. (11) can lead to a loss of accuracy. To avoid this, we find $\mathcal{R}_{1J_n,1}(q\omega, 2q\omega, q\omega)$, solving again the inhomogeneous equation (9) with $|\delta\phi_q\rangle$ (instead of $|0\rangle$) on the right-hand side.

The calculation of the terms $\mathcal{R}'_{101}(q\omega, 0, q\omega)$ is carried out in a similar way. For example, for $\mathcal{R}'_{101}(\omega, 0, \omega)$ we arrive at

$$\mathcal{R}'_{101}(\omega, 0, \omega) = \sum_{\gamma_n}' \frac{|\langle \delta\phi_{+1} \| D \| \gamma_n, 0 \rangle|^2}{E_n - E_0}. \quad (12)$$

However, there is a complication in comparison to the previous case: we need to exclude the term $|\gamma_n, 0\rangle = |\gamma_0, 0\rangle$ from the summation over γ_n . We developed the following method to eliminate this term from the solution of the inhomogeneous equation. We add a small imaginary term $i\omega'$ to the denominator of Eq. (12), so that

$$\mathcal{R}'_{101}(\omega, 0, \omega) = \text{Re}\{\mathcal{R}'_{101}(\omega, i\omega' \rightarrow 0, \omega)\},$$

and find the real part of $\mathcal{R}'_{101}(\omega, i\omega', \omega)$, given by

$$\text{Re}\{\mathcal{R}'_{101}(\omega, i\omega', \omega)\} = \sum_{\gamma_n} \frac{(E_n - E_0) |\langle \delta\phi_{+1} \| D \| \gamma_n, 0 \rangle|^2}{(E_n - E_0)^2 + (\omega')^2}.$$

The term $|\gamma_n, 0\rangle = |\gamma_0, 0\rangle$ automatically disappears because of the $(E_n - E_0)$ in the numerator.

In practice, the parameter ω' should be chosen to be sufficiently small to satisfy the condition $|\omega'| \ll |E_0 - E_n|$ for any n , but preserve numerical stability when solving the inhomogeneous equation at this ω' . To ensure more flexibility in choosing the parameter ω' , we orthogonalize $|\delta\phi_q\rangle$ to $|0\rangle$, as

$$|\delta\phi_q\rangle' = (1 - |0\rangle\langle 0|)|\delta\phi_q\rangle,$$

and use $|\delta\phi_q\rangle'$ instead of $|\delta\phi_q\rangle$.

The term $\mathcal{R}_1(q\omega) = (3/2)\alpha_{q\omega}$ is calculated following Eqs. (8)–(10), but the quantity $\mathcal{R}_1(\omega, \omega)$ has an additional energy denominator. Therefore, we treat this term as the derivative of $\mathcal{R}_1(\omega)$ over ω , i.e.,

$$\mathcal{R}_1(\omega, \omega) = \frac{\partial \mathcal{R}_1(\omega)}{\partial \omega} = \frac{3}{2} \lim_{\Delta \rightarrow 0} \frac{\alpha_{\omega+\Delta} - \alpha_{\omega}}{\Delta}.$$

We find $\mathcal{R}_1(\omega)$ and $\mathcal{R}_1(\omega, \omega)$ by calculating α_{ω} and $\alpha_{\omega+\Delta}$, where Δ is chosen so that $\Delta \ll \omega$.

Method: Sr calculations.—The electric dipole polarizabilities of the Sr clock states at the magic wavelength $\lambda^* = 813.4280(5)$ nm [26] were calculated in our work [27] to be $\alpha_{E1}(\omega^*) = 286.0(3)$ a.u..

In this work we apply the method discussed above to calculate the hyperpolarizabilities and the multipolar polarizabilities, α^{M1} and α^{E2} , for the Sr clock states, $5s^2\ ^1S_0$ and $5s5p\ ^3P_0^o$, at the magic frequency. We consider Sr as an atom with two valence electrons above the closed shell core. The calculations are carried out in the framework of the high-precision relativistic methods combining configuration interaction (CI) with (i) many-body perturbation theory [28] and (ii) the linearized coupled-cluster method [29]. In these methods, the energies and wave functions are found from the multiparticle Schrödinger equation

$$H_{\text{eff}}(E_n)\Phi_n = E_n\Phi_n, \quad (13)$$

where the effective Hamiltonian is defined as

$$H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E). \quad (14)$$

Here, H_{FC} is the Hamiltonian in the frozen core (Dirac-Hatree-Fock) approximation and Σ is the energy-dependent correction, which takes into account virtual core excitations in the second order of the perturbation theory (the CI + MBPT method) or to all orders (the CI + all-order method).

Hyperpolarizabilities.—The hyperpolarizability is dominated by the valence electrons' contribution; the contribution of the core electrons is small and can be neglected at

the present level of accuracy. In particular, the static hyperpolarizability of the Sr^{2+} ionic core in the ground state was found in Ref. [30] to be negligible, 62.6 a.u.

To accurately calculate the valence part of the quantities given by Eqs. (6) and (7), we applied solutions of the inhomogeneous equations as was described above. We use effective (or “dressed”) operators in our calculations that include the random-phase approximation (RPA). A concept of effective operators was developed in Ref. [31].

The breakdown of the terms contributing to 1S_0 and ${}^3P_0^o$ hyperpolarizabilities at the magic frequency is given in Table I. We carry out all calculations using both the CI + MBPT and CI + all-order methods. The CI + all-order calculations include higher-order terms in comparison with the CI + MBPT calculations and are more accurate. The difference between these two calculations gives an estimate of the uncertainty of the results. This method in evaluating uncertainties has been extensively tested for Sr and other atoms [27,32,33].

We compare our recommended value for the differential clock hyperpolarizability at the magic frequency,

TABLE I. Contributions to the Sr hyperpolarizabilities $\beta(5s^2 {}^1S_0)$ and $\beta(5s5p {}^3P_0^o)$ (in a.u.) calculated in the CI + all-order (labeled as “CI + all”) and CI + MBPT (labeled as “CI + PT”) approximations at the magic frequency. $\Delta\beta \equiv \beta({}^3P_0^o) - \beta({}^1S_0)$ is the difference of the total ${}^3P_0^o$ and 1S_0 values. Numbers in brackets represent powers of 10. The uncertainties are given in parentheses.

Contribution	$5s^2 {}^1S_0$		$5s5p {}^3P_0^o$	
	CI + All	CI + PT	CI + All	CI + PT
$\frac{1}{9}\mathcal{R}_{101}(\omega, 2\omega, \omega)$	5.08[5]	4.62[5]	-5.96[6]	-5.64[6]
$\frac{1}{9}\mathcal{R}_{101}(-\omega, -2\omega, -\omega)$	4.48[4]	4.38[4]	9.83[4]	9.91[4]
$\frac{1}{9}\mathcal{R}'_{101}(\omega, 0, \omega)$	2.41[5]	2.29[5]	2.43[6]	2.68[6]
$\frac{1}{9}\mathcal{R}'_{101}(\omega, 0, -\omega)$	6.47[4]	6.20[4]	3.26[5]	3.47[5]
$\frac{1}{9}\mathcal{R}'_{101}(-\omega, 0, \omega)$	6.47[4]	6.20[4]	3.26[5]	3.47[5]
$\frac{1}{9}\mathcal{R}'_{101}(-\omega, 0, -\omega)$	1.76[4]	1.70[4]	6.78[4]	6.84[4]
$\mathcal{R}_1(\omega)$	6.63[2]	6.49[2]	6.03[2]	6.39[2]
$\mathcal{R}_1(-\omega)$	1.94[2]	1.92[2]	2.67[2]	2.68[2]
$\mathcal{R}_1(\omega, \omega)$	1.52[4]	1.47[4]	4.79[4]	5.12[4]
$\mathcal{R}_1(-\omega, -\omega)$	1.17[3]	1.17[3]	2.40[3]	2.40[3]
$\frac{1}{9}Y_{101}(\omega)$	-6.18[5]	-6.06[5]	-7.57[6]	-7.50[6]
$\frac{2}{45}\mathcal{R}_{121}(\omega, 2\omega, \omega)$	7.61[5]	7.10[5]	-1.82[7]	-1.65[7]
$\frac{2}{45}\mathcal{R}_{121}(-\omega, -2\omega, -\omega)$	1.83[4]	1.78[4]	4.97[4]	5.03[4]
$\frac{2}{45}\mathcal{R}_{121}(\omega, 0, \omega)$	3.86[5]	3.68[5]	9.64[6]	1.13[7]
$\frac{2}{45}\mathcal{R}_{121}(\omega, 0, -\omega)$	1.07[5]	1.03[5]	8.72[5]	9.76[5]
$\frac{2}{45}\mathcal{R}_{121}(-\omega, 0, \omega)$	1.07[5]	1.03[5]	8.72[5]	9.76[5]
$\frac{2}{45}\mathcal{R}_{121}(-\omega, 0, -\omega)$	2.98[4]	2.90[4]	1.44[5]	1.50[5]
$\frac{2}{45}Y_{121}(\omega)$	1.41[6]	1.33[6]	-6.58[6]	-3.03[6]
Total	7.90[5]	7.25[5]	-1.42[7]	-1.05[7]
Recommended	7.90(65)[5]		-1.42(37)[7]	
$\Delta\beta$	-1.50[7]	-1.12[7]		
$\Delta\beta$ (Recommended)	-1.5(4)[7]			

$\Delta\beta = \beta({}^3P_0^o) - \beta({}^1S_0)$, with the experimental results of the JILA and SYRTE groups, and also with the theoretical value obtained in Ref. [19]:

$$\begin{aligned} \Delta\beta &= -1.5(4) \times 10^7 \text{ a.u. (this work),} \\ &= -2.01(45) \times 10^7 \text{ a.u. (SYRTE[21, 22]),} \\ &= -1.3(1.3) \times 10^7 \text{ a.u. (JILA[3]),} \\ &= -3.74 \times 10^7 \text{ a.u. (theory[19]).} \end{aligned}$$

Our result is in agreement with the experimental results, but definitely not consistent with zero.

M1 and E2 polarizabilities at the magic frequency.—To accurately calculate the valence part of the $E2$ polarizabilities of the clock states at the magic frequency, given by Eq. (4), we solved an inhomogeneous equation similar to Eq. (9), as described above, with the electric quadrupole operator Q . We calculated these quantities using both the CI + all-order and CI + MBPT methods, including the RPA corrections to the operator Q . The core contributions were calculated in the RPA. For the $M1$ polarizabilities, only a few low-lying intermediate states give dominant contributions, and it is sufficient to calculate their sum. We estimate the uncertainties of the results as the difference between the CI + all-order and CI + MBPT values.

The $\alpha^{M1}({}^1S_0)$ polarizability is very small and can be neglected. The $\alpha^{M1}({}^3P_0^o)$ polarizability is more than 3 orders of magnitude larger, but still an order of magnitude smaller than $\Delta\alpha^{E2}$. Therefore, the accuracy of $\Delta\alpha^{QM} \equiv \Delta\alpha^{E2} + \Delta\alpha^{M1}$, where

$$\begin{aligned} \Delta\alpha^{M1} &\equiv \alpha^{M1}({}^3P_0^o) - \alpha^{M1}({}^1S_0), \\ \Delta\alpha^{E2} &\equiv \alpha^{E2}({}^3P_0^o) - \alpha^{E2}({}^1S_0), \end{aligned} \quad (15)$$

is mostly determined by the uncertainty in $\Delta\alpha_{E2}$.

TABLE II. The dynamic $M1$ and $E2$ polarizabilities (in a.u.) of the $5s^2 {}^1S_0$ and $5s5p {}^3P_0^o$ states at the magic frequency, calculated in the CI + MBPT (labeled as “CI + PT”) and CI + all-order (labeled as “CI + all”) approximations. The recommended value of $\Delta\alpha^{QM}$ is given in the last line. The uncertainties are given in parentheses.

Polariz.	CI + PT	CI + All	Other theor.
$\alpha^{M1}({}^1S_0)$	2.19×10^{-9}	2.37×10^{-9}	
$\alpha^{M1}({}^3P_0^o)$	-5.09×10^{-6}	-5.08×10^{-6}	
$\Delta\alpha^{M1}$	-5.09×10^{-6}	-5.08×10^{-6}	
$\alpha^{E2}({}^1S_0)$	8.61×10^{-5}	$8.87(26) \times 10^{-5}$	
$\alpha^{E2}({}^3P_0^o)$	12.1×10^{-5}	$12.2(25) \times 10^{-5}$	
$\Delta\alpha_{E2}$	3.50×10^{-5}	$3.31(36) \times 10^{-5}$	
$\Delta\alpha^{QM}$	2.99×10^{-5}	$2.80(36) \times 10^{-5}$	0.74×10^{-5} [19]
Rec. $\Delta\alpha^{QM}$		$2.80(36) \times 10^{-5}$	-3.60×10^{-5} [18]

The final values of the polarizabilities, the recommended value of $\Delta\alpha^{QM}$, and their uncertainties are listed in Table II. We estimate the uncertainty of the recommended value of $\Delta\alpha^{QM}$ to be 13%.

To conclude, we have developed a method to calculate hyperpolarizabilities for atoms and ions based on a solution of inhomogeneous equation (9), which allows us to carry out complete summations over all intermediate states. We applied our method to the calculation of the hyperpolarizabilities for the 1S_0 and $^3P_0^o$ clock states in Sr and found the differential hyperpolarizability to be $-1.5(4) \times 10^7$ a.u. We have also calculated the $M1$ and $E2$ polarizabilities for these states at the magic frequency.

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