Calculation of higher-order corrections to the light shift of the $5s^2\,^1S_0 - 5s5p\,^3P_0^o$ clock transition in Cd

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In the recent work by Yamaguchi et al. [Phys. Rev. Lett. 123, 113201 (2019)] Cd was identified as an excellent candidate for a lattice clock. Here, we carried out computations needed for further clock development and made an assessment of the higher-order corrections to the light shift of the $5s^2\,^1S_0 - 5s5p\,^3P_0^o$ clock transition. We carried out calculations of the magnetic dipole and electric quadrupole polarizabilities and linear and circular hyperpolarizabilities of the $5s^2\,^1S_0$ and $5s5p\,^3P_0^o$ clock states at the magic wavelength and estimated uncertainties of these quantities. We also evaluated the second-order Zeeman clock transition frequency shift.

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I. INTRODUCTION

The Cd $5s^2\,^1S_0 - 5s5p\,^3P_0^o$ transition has several desirable attributes for the development of a lattice clock. This clock has more than an order of magnitude smaller blackbody radiation (BBR) shift (a Stark shift resulting from the thermal radiation of the atoms) in comparison with Sr and Yb [1–3]. The size of a BBR shift is a property of the specific atomic transition used as a frequency standard and an uncertainty in the BBR shift is known to be one of the limiting systematic uncertainties in the clock uncertainty budget [4,5]. Short of cryogenic cooling, it cannot be suppressed and needs to be quantified with high accuracy.

Two isotopes, $^{111}$Cd and $^{113}$Cd, both with 12% natural abundance, have a nuclear spin of 1/2, which precludes tensor light shifts from the lattice light, another advantageous feature. Cd has the narrow $5s^2\,^1S_0 - 5s5p\,^3P_0^o$ intercombination transition, allowing Doppler cooling to 1.58 $\mu$K and simplifying control of higher-order lattice light shifts [1]. The light for all of the transitions needed for the Cd clock, including the magic lattice, can be generated by direct or frequency-doubled or -quadrupled semiconductor lasers [1].

In 2019, the Cd clock magic wavelength was measured to be 419.88(14) nm [1], in excellent agreement with a theoretical calculation reported in the same work. At the magic wavelength, upper and lower clock states experience the same light shift, up to multipolar and higher-order effects considered in this work. The fractional BBR shift was calculated to be 2.83(8) $\times$ 10$^{-16}$ at 300 K in Ref. [1], in agreement with Ref. [3]. Recent progress opens a pathway to rapid progress in Cd clock development and calls for a detailed investigation of the clock systematic effects.

In this work we calculated properties needed to quantify higher-order light shifts: magnetic dipole and electric quadrupole polarizabilities and linear and circular hyperpolarizabilities of the $5s^2\,^1S_0$ and $5s5p\,^3P_0^o$ clock states at the magic wavelength, and estimated uncertainties of these quantities. We also evaluated the second-order Zeeman clock transition frequency shift in the presence of a weak magnetic field.

The paper is organized as follows. The general formalism and main formulas are presented in Sec. II. In Sec. III, we briefly describe the method of calculation. Section IV is devoted to a discussion of the results obtained, and Sec. V contains concluding remarks.

II. GENERAL FORMALISM

We consider the Cd atom in a state $|$0$\rangle$ (with the total angular momentum $J = 0$) placed in a field of the lattice standing wave with the electric-field vector given by

$$\mathbf{E} = 2\mathbf{E}_0 \cos(kx) \cos(\omega t).$$

(1)

Here $k = \omega/c$, $\omega$ 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. The atom-lattice interaction leads to the optical lattice potential for the atom that at $|kx| \ll 1$ can be approximated as [2,6]

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

(2)

Here $\alpha^{E1}$, $\alpha^{M1}$, and $\alpha^{E2}$ are the electric dipole, magnetic dipole, and electric quadrupole polarizabilities, respectively, and $\beta$ is the hyperpolarizability defined below.

The $\alpha 2K$-pole polarizability of the $|0\rangle$ state with the energy $E_0$ is expressed (we use atomic units $\hbar = m = |e| = 1$) as [7]

$$\alpha^{2K}(\omega) = \frac{K + 1}{K} \frac{2K + 1}{[(2K + 1)!]^2(\omega \alpha)^{2K-2}} \sum_n \frac{(E_n - E_0)(n||T_{2K}||0)^2}{(E_n - E_0)^2 - \omega^2},$$

(3)
where \( \lambda \) stands for electric, \( \lambda = E \), and magnetic, \( \lambda = M \), multipoles and \( |n|R_{k}\rangle = 0 \) are the reduced matrix elements of the multipole operators, \( T_{k} = D, T_{M} = \mu \), and \( T_{E} = Q \).

The expression for the hyperpolarizability of the \(|0\rangle\) state depends on the polarization of the lattice wave. Below we consider the cases when the lattice wave is linearly or circularly polarized, and the fourth-order correction to an atomic energy is determined by the linear or circular hyperpolarizability, respectively.

The expression for the linear hyperpolarizability \( \beta_{l}(\omega) \) is given by [6]

\[
\beta_{l}(\omega) = \frac{1}{9} Y_{101}(\omega) + \frac{2}{45} Y_{121}(\omega),
\]
with the quantities \( Y_{101}(\omega) \) and \( Y_{121}(\omega) \) determined as

\[
Y_{101}(\omega) = \sum_{q} \mathcal{R}_{101}(q\omega, 2q\omega, q\omega) + \sum_{q, q'} \mathcal{R}_{101}'(q\omega, 0, q'\omega) - \mathcal{R}_{1}(q'\omega)\mathcal{R}_{1}(q\omega, q\omega),
\]

\[
Y_{121}(\omega) = \sum_{q} \left( \gamma_{q} J_{0} \langle d|\gamma_{m} J_{m}\rangle \langle d|\gamma_{n} J_{n}\rangle \langle d|\gamma_{k} J_{k}\rangle \langle \gamma_{l} J_{l}|d|\gamma_{0} J_{0}\rangle \right)\left( E_{m} - E_{0} - \omega_{1} E_{n} - E_{0} - \omega_{2} E_{k} - E_{0} - \omega_{3} \right),
\]

\[
\mathcal{R}_{101}(\omega, \omega, \omega) = \sum_{\gamma_{0}, \gamma_{e}} \frac{E_{m} - E_{0} - \omega_{1}}{E_{m} - E_{0} - \omega_{2}} \langle \gamma_{e} J_{0}|d|\gamma_{m} J_{m}\rangle \langle \gamma_{e} J_{0}|d|\gamma_{n} J_{n}\rangle \langle \gamma_{e} J_{0}|d|\gamma_{k} J_{k}\rangle \langle \gamma_{l} J_{l}|d|\gamma_{0} J_{0}\rangle ^{2},
\]

\[
\mathcal{R}_{101}(\omega, \omega, \omega) = \sum_{\gamma_{0}, \gamma_{e}} \frac{E_{m} - E_{0} - \omega_{1}}{E_{m} - E_{0} - \omega_{2}} \langle \gamma_{e} J_{0}|d|\gamma_{m} J_{m}\rangle \langle \gamma_{e} J_{0}|d|\gamma_{n} J_{n}\rangle \langle \gamma_{e} J_{0}|d|\gamma_{k} J_{k}\rangle \langle \gamma_{l} J_{l}|d|\gamma_{0} J_{0}\rangle ^{2}.
\]

The notation \( \mathcal{R}_{101} \), i.e., the prime over \( \mathcal{R} \), means that the term \( |\gamma_{e} J_{0}|d|\gamma_{m} J_{m}\rangle \langle d|\gamma_{n} J_{n}\rangle \langle d|\gamma_{k} J_{k}\rangle \langle \gamma_{l} J_{l}|d|\gamma_{0} J_{0}\rangle \) should be excluded from the summation over \( \gamma_{0} \) in Eq. (6).

The properties of the lattice potential for the Cd atom in its ground and excited clock states are determined by Eq. (2) and depend on the frequency. Below we analyze these properties at the experimentally determined magic wavelength \( \lambda^{*} = 419.88(14) \) nm [1]. The magic frequency, \( \omega^{*} \), corresponding to this wavelength, is \( \omega^{*} \approx 23816 \) cm\(^{-1} \) \( \approx 0.108515 \) a.u.

At the magic frequency the electric dipole polarizabilities of the clock \( 5s^{2}\uparrow S_{0} \) and \( 5s^{5}p^{3}\downarrow P_{0} \) states are equal to each other, i.e., \( \alpha_{0}^{S_{0}}(\omega^{*}) = \alpha_{0}^{P_{0}}(\omega^{*}) \). These polarizabilities were calculated in Ref. [11] and carry out accurately three summations over intermediate states, we solve the inhomogeneous equation twice. A detailed description of this approach is given in Ref. [6].

Using the formulas given above, we calculated the \( M1 \) and \( E2 \) polarizabilities and the linear and circular hyperpolarizabilities \( \beta_{l,c} \) of the clock states at the magic frequency \( \omega^{*} \), found respective differential polarizabilities and hyperpolarizabilities, and determined uncertainties of these values.

### III. METHOD OF CALCULATION

We carried out calculations in the framework of high-accuracy relativistic methods combining configuration interaction (CI) with (i) many-body perturbation theory (CI + MBPT method [8]) and (ii) the linearized coupled-cluster approach (CI + all-order method) [9]. In these methods the energies and wave functions are found from the multiparticle Schrödinger equation

\[
H_{\text{eff}}(E)\Phi_{n} = E_{n}\Phi_{n},
\]

where the effective Hamiltonian is defined as

\[
H_{\text{eff}}(E) = H_{\text{FC}} + \Sigma(E).
\]

Here, \( H_{\text{FC}} \) is the Hamiltonian in the frozen core approximation and \( \Sigma \) 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 in all orders of the perturbation theory (the CI + all-order method).

To accurately calculate the valence parts of the polarizabilities and hyperpolarizabilities, we solve the inhomogeneous equation using the Sternheimer [10] or Dalgarno-Lewis [11] method following the formalism developed in Ref. [12]. We use an effective (or "dressed") electric-dipole operator in our calculations that includes the random-phase approximation (RPA). To calculate such complicated quantities as \( \mathcal{R}_{101} \) and carry out accurately three summations over intermediate states, we solve the inhomogeneous equation twice. A detailed description of this approach is given in Ref. [6].

### IV. RESULTS AND DISCUSSION

We carried out calculations of the \( M1 \) and \( E2 \) polarizabilities and the hyperpolarizabilities in the CI + MBPT and CI + all-order approximations. In both cases the theoretical energies were used. The CI + all-order calculations include higher-order terms in comparison with the CI + MBPT calculations and are more accurate. The difference of these two calculations gives us an estimate of the uncertainty of
the results. This method in evaluating uncertainties has been extensively tested in calculating clock-related properties (including hyperpolarizabilities) of other divalent elements such as Sr [6,13], Lu+ [14], and Yb [15].

A. Linear and circular hyperpolarizabilities of the $^1S_0$ and $^3P_0^0$ clock states

In calculating quantities given by Eqs. (6) and (7) a main contribution comes from valence electrons. The core electrons contribution is much smaller and we included it only to $R_1(\omega)$ terms. As seen from Eq. (7), $R_1(\omega)$ can be treated as the quantity proportional to the scalar static polarizability of the $|\gamma_0\gamma_0\rangle$ state with the energy $E_0$ shifted as $E_0 + \omega$. A breakdown of different contributions (including core contribution) to the static polarizabilities of the clock states with estimates of their uncertainties is provided in Ref. [1].

As follows from Eq. (7), the quantity $R_1(\omega, \omega)$ can be treated as the derivative of $R_1(\omega)$ over $\omega$, i.e.,

$$R_1(\omega, \omega) \equiv \frac{\partial R_1(\omega)}{\partial \omega} = \lim_{\Delta \to 0} \frac{R_1(\omega + \Delta) - R_1(\omega)}{\Delta}.$$ 

Since the core contribution to $R_1(\omega)$ is rather insensitive to $\omega$ and $\Delta$ is small, the core contributions to $R_1(\omega + \Delta)$ and $R_1(\omega, \omega)$ are practically identical and cancel each other in the expression for $R_1(\omega, \omega)$.

Taking into account the uncertainty of our results for the $^1S_0$ and $^3P_0^0$ hyperpolarizabilities, we assume that the core contribution to the $R_{1\mu}(\omega_1, \omega_2, \omega_3)$ terms is also negligible. This assumption is based on the calculation of the static hyperpolarizability for the $Sr^{2+}$ ground state that was found to be 62.6 a.u. [16]. This is negligibly small compared to the valence contribution to $R_{1\mu}(\omega_1, \omega_2, \omega_3)$ in the case of the quite similar $5s^2$ $^1S_0$ and $5s5p$ $^3P_0^0$ clock states in Sr [6].

The results of calculation of the linear and circular hyperpolarizabilities of the $^1S_0$ and $^3P_0^0$ clock states are presented in Table I. Our recommended value of differential linear hyperpolarizability, $\Delta \beta_i(\omega^*) = -1.85(50) \times 10^5$ a.u., is two orders of magnitude smaller (in absolute value) than the analogous differential hyperpolarizability for Sr, $\Delta \beta_i = -1.5(4) \times 10^7$ a.u. [6]. In the case of Cd, the absolute values of the contributing terms are generally smaller than in Sr, and there are significant cancellations between them.

The circular hyperpolarizability of the $^3P_0^0$ state is two orders of magnitude larger in absolute value than the circular hyperpolarizability of the $^1S_0$ state and the linear hyperpolarizability of the $^3P_0^0$ state. This is explained as follows: the main contribution to $\beta_i(\gamma_0^\alpha)(\omega^*)$ comes from the term

$$\gamma_0^\alpha E_{m^\alpha} - \omega^* \gamma_0^\alpha E_{m^\alpha - \omega^*}$$

In the sum over $\gamma_0^\alpha$ there is the intermediate state 5s5p $^3P_0^0$ separated from $^3P_0^0$ by the fine-structure interval. In this case the energy denominator $E_{m^\alpha} - E_{m^\alpha - \omega^*} \approx 542$ cm$^{-1}$ is small and, consequently, the contribution of this term is large, leading to much larger hyperpolarizability for the circular polarization.

We compare our results with those obtained in Ref. [2] in Table I. There is a reasonable agreement for differential linear hyperpolarizability while our differential linear hyperpolarizability is five times smaller in absolute value than that found in Ref. [2].

B. $M1$ 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, we solved inhomogeneous equation with the electric quadrupole operator $Q$ in the right-hand side. As in the case of hyperpolarizability, 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 final values of the polarizabilities and their uncertainties are listed in Table II. We also determined the recommended value of $\Delta \alpha'^M = \Delta \alpha'^{E2} + \Delta \alpha'^{M1}$, where

$$\Delta \alpha'^{M1} \equiv \alpha'^{M1}(\gamma_0^\alpha) - \alpha'^{M1}(^1S_0),$$

$$\Delta \alpha'^{E2} \equiv \alpha'^{E2}(\gamma_0^\alpha) - \alpha'^{E2}(^1S_0).$$

To determine the uncertainty of $\Delta \alpha'^M$ we note that the $\alpha'^{M1}(^1S_0)$ polarizability is very small and we can neglect it. The $\alpha'^{M1}(\gamma_0^\alpha)$ polarizability is more than three orders of magnitude smaller in absolute value than $\alpha'^{M1}(^1S_0)$, but still an order of magnitude smaller than $\Delta \alpha'^{E2}$. Therefore, the uncertainty of $\Delta \alpha'^M$ is mostly determined by the uncertainty in $\Delta \alpha'^{E2}$ and we estimate it to be 4%. Comparing our recommended value for $\Delta \alpha'^M$ with the result obtained in Ref. [2], we see that there is a fair agreement between them.

C. Second-order Zeeman shift

In this section we consider a systematic effect due to second-order Zeeman shift which both clock states experience in the presence of a weak uniform external magnetic field. If an atom is placed in a such magnetic field $B$, the interaction Hamiltonian can be written as [17] (we use the cgs system of units),

$$H = -\mu \cdot B + \frac{e^2}{8mc^2} \sum_{i=1}^{Z} (\mathbf{B} \times \mathbf{r}_i)^2,$$

where $(\mathbf{B} \times \mathbf{r}_i)$ is the vector product of $\mathbf{B}$ and $\mathbf{r}_i$. The atomic magnetic moment $\mu$ is mostly determined by the electronic
TABLE I. Contributions to the linear and circular hyperpolarizabilities \( \beta_{\alpha}(5s^2 \, 1S_0) \) and \( \beta_{\alpha}(5s5p \, 3p_0) \) (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 \( \omega^* = 0.108 \times 515 \) a.u. The "Total" values are obtained according to Eqs. (4) and (5). \( \Delta \beta_{\alpha} = \beta_{\alpha}(5s^2 \, 1S_0) - \beta_{\alpha}(5s5p \, 3p_0) \) is the difference of the "Total" \( 3p_0 \) and \( 1S_0 \) values. Numbers in brackets represent powers of 10. The uncertainties are given in parentheses.

![Table I](image)

The magnetic moment and is given by

\[
\mu = -\mu_0(J + S),
\]

where \( J \) and \( S \) are the total and spin angular momenta of the atomic state and \( \mu_0 \) is the Bohr magneton defined as \( \mu_0 = e\hbar/(2mc) \). Here \( e \) and \( m \) are the electron charge and mass, \( c \) is the speed of light, and \( \hbar \) is the Plank constant. In the second term of Eq. (11) the summation goes over all electrons in the atom and the vector potential \( A \) is chosen in the form \( A(r) = \frac{1}{2}(B \times \mathbf{r}) \).

Directing the external magnetic field \( B \) along the z-axis (\( B = B_z \equiv B \)), we can find the second-order Zeeman shift, \( \Delta E^{(1)} \), due to the first term in Eq. (11) (in the absence of hyperfine interaction) as

\[
\Delta E^{(1)} = -\frac{1}{2} \alpha^{M1} B^2,
\]

where \( \alpha^{M1} \) is the magnetic-dipole polarizability. For a state \( |J = 0\rangle \) it is reduced to the scalar polarizability, given by

\[
\alpha^{M1} = \frac{2}{3} \sum_n \frac{|\langle n | \mu | J = 0 \rangle|^2}{E_n - E_0}.
\]

To estimate the second-order Zeeman shift for the clock transition

\[
\Delta \nu^{(1)} = \frac{\Delta E^{(1)}(3p_0) - \Delta E^{(1)}(1S_0)}{\hbar},
\]

we note that the \( \alpha^{M1}(1S_0) \) polarizability is negligibly small compared to \( \alpha^{M1}(3p_0) \), so we can write \( \Delta \nu^{(1)} = \Delta E^{(1)}(3p_0)/\hbar \).

For an estimate of \( \alpha^{M1}(3p_0) \) we take into account that the main contribution to this polarizability comes from the intermediate state \( 5s5p \, 3p_0 \). Then, from Eq. (14) we obtain

\[
\alpha^{M1}(3p_0) \approx \frac{2}{3} \frac{\langle 3p_0 | |\mu| | 3p_0 \rangle^2}{E_{3p_0} - E_{3p_0} - B^2},
\]

The matrix element \( \langle 3p_0 | |\mu| | 3p_0 \rangle \) can be found if we note that the total angular momentum operator \( J \) does not mix the states with different \( J \), while the operator \( S \) acts only on the spin variables and its matrix element can be calculated using an analytical formula valid in the \((LSJM)\) representation [18]. Then, we obtain

\[
\langle 3p_0 | |\mu| | 3p_0 \rangle = -\mu_0 \delta_{p_0} (|S|) |3p_0 \rangle = \sqrt{2} \mu_0.
\]

We note that the numerical calculation of this matrix element gives the value 1.412\( \mu_0 \), which is very close to the analytical result.

Substituting it into Eq. (13) we find

\[
\Delta E^{(1)}(3p_0) \approx \frac{2}{3} \frac{\mu_0^2}{E_{3p_0} - E_{3p_0} - B^2},
\]

in agreement with the result obtained in Ref. [19].

Using the experimental value of the energy difference \( E_{3p_0} - E_{3p_0} \approx 542 \) cm\(^{-1} \), we arrive at

\[
\Delta \nu^{(1)} \approx -80 B^2,
\]

where \( \Delta \nu^{(1)} \) is in mHz and the magnetic field \( B \) is in G.

Due to smallness of the energy difference between the fine-structure levels in Eq. (16), the first term in Eq. (11) usually gives a larger contribution to the second-order Zeeman shift than the second term [17]. To estimate the contribution of the second term we note that

\[
(B \times \mathbf{r})^2 = B^2 r^2 \sin^2 \theta \equiv B^2 (r^2 - r_z^2),
\]

where we omitted the subscript \( i \) for brevity. Forming the \( z \) component of the second-order rank tensor \( R \) as \( R_{zz} = R_{z0} = r_z^2 - r^2/3 \) and taking into account that, according to

\[
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\]

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the Wigner-Eckart theorem, \( \langle 0 | R_{20} | 0 \rangle = 0 \) for a state \( |0, J = 0\rangle \), we have

\[
\langle 0 | r_i^2 | 0 \rangle = \frac{1}{3} \langle 0 | r_i^2 | 0 \rangle \tag{18}
\]

arriving at

\[
\Delta E^{(2)} = \frac{e^2}{8mc^2} \sum_{i=1}^{Z} (| \mathbf{B} \times \mathbf{r}_i |^2 | 0 \rangle \tag{19}
\]

\[
\approx \frac{e^2 B^2}{12mc^2} \sum_{i=1}^{Z} r_i^2 | 0 \rangle .
\]

The respective contribution to the second-order Zeeman shift of the clock transition is

\[
\Delta \nu^{(2)} = \frac{e^2 B^2}{12mc^2} \sum_{i=1}^{Z} \left\{ \langle 3p^0_0 | r_i^2 | 3p^0_0 \rangle - \langle 1s_0 | r_i^2 | 1s_0 \rangle \right\} . \tag{20}
\]

To carry out summation over all atomic electrons and estimate the matrix elements in Eq. (20) we used the CI method, in which all 48 electrons were placed in the valence field and several most important configurations were included in consideration. According to our estimate,

\[
\sum_{i=1}^{Z} \left\{ \langle 3p^0_0 | r_i^2 | 3p^0_0 \rangle - \langle 1s_0 | r_i^2 | 1s_0 \rangle \right\} \approx 7 a_0^2 . \tag{21}
\]

(where \( a_0 \) is the Bohr radius) and finally \( \Delta \nu^{(2)} [\text{mHz}] \approx 0.7 B^2 \) (with the magnetic field \( B \) is in G). Thus, \( \Delta \nu^{(2)} \) is two orders of magnitude smaller in absolute value than \( \Delta \nu^{(1)} \).

V. CONCLUSION

We carried out calculations of the magnetic dipole and electric quadrupole polarizabilities as well as linear and circular hyperpolarizabilities of the clock \( 5s^2 \, ^1S_0 \) and \( 5s5p \, ^3P^0_0 \) states at the magic wavelength and compared them with other available data. We also evaluated the second-order Zeeman shift for the clock transition frequency. These values are required for an assessment of the higher-order corrections to the light shift of the \( 5s^2 \, ^1S_0 - 5s5p \, ^3P^0_0 \) clock transition. We have demonstrated that the linear differential hyperpolarizability for the clock transition for Cd is two orders of magnitude smaller than for Sr and Yb. We also found the circular hyperpolarizability to be much larger than the linear hyperpolarizability and explained the source of this difference. Knowledge of the multipolar polarizabilities and hyperpolarizabilities at different polarizations of the lattice wave is needed for further Cd clock development and selection of the lattice configurations to minimize the higher-order light shifts.

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