Magic Wavelengths for Terahertz Clock Transitions

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Magic wavelengths for laser trapping of boson isotopes of alkaline-earth Sr, Ca and Mg atoms are investigated while considering terahertz clock transitions between the \(^3P_0, ^3P_1, ^3P_2\) metastable triplet states. Our calculation shows that magic wavelengths of trapping laser do exist. This result is important because these metastable states have already been used to realize accurate clocks in the terahertz frequency domain. Detailed discussions for magic wavelength for terahertz clock transitions are given in this paper.

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I. INTRODUCTION

Frequency standards have achieved an unprecedented success in experimental demonstrating accuracies of \(4 \times 10^{-16}\) with a cesium microwave fountain clock [1] and \(1.9 \times 10^{-17}\) with an ion optical clock [2, 3]. For optical frequency standards based on neutral atoms, in order to effectively increase the interrogation time, Katori proposed to utilize optical lattice trap formed with a magic wavelength trapping laser [4, 5]. This clever technique greatly enhanced established high-accuracy optical frequency standard with neutral Sr atom to an accuracy of \(1 \times 10^{-16}\) [6-8]. Different optical clock schemes based on Ca [9], Yb [10, 11] atoms trapped with magic wavelength lasers have been proposed.

Optical trap with a far off-resonant laser is a very useful tool for the confinement of cold atoms. Nevertheless, for the precision of clock transitions in frequency standards, light shift due to trapping laser has to be avoided. Thus the wavelength of the trapping laser should be tuned to a region where the light shift for the clock transition is eliminated, that means the light shifts of the two clock transition states cancel each other. The wavelength \(\lambda\) is called magic wavelength [4, 5]. Recently, cesium primary frequency standard with atoms trapped in an optical lattice with a magic wavelength was suggested [12, 13], and possible magic wavelengths for clock transitions in aluminium and gallium atoms were also calculated [14].

In contrast to the above mentioned magic wavelengths for optical clock transitions and microwave clock transitions, here we investigate magic wavelengths for terahertz clock transitions. Absolute frequency standards in the terahertz domain with fine structure transition lines of the Mg and Ca metastable triplet states were first proposed in 1972 by Struman [15]. After more than twenty years of continuing improvement, a frequency standard based on the \(^3P_1, ^3P_0\) Mg transition and thermal atoms in a beam has reached an uncertainty of \(1 \times 10^{-12}\) [16, 17]. However, these potential terahertz transitions for high-resolution clock references have never been experimentally investigated with laser cooled or laser trapped atoms.

In this paper, we present our most recent calculation of trapping laser magic wavelengths for Sr, Ca and Mg atoms, considering different possible clock transitions between metastable triplet states \(^3P\). Accurate terahertz clocks could then be built based on such atoms which are cooled and trapped in an optical lattice.

II. THEORETICAL DESCRIPTION

For alkaline-earth atoms, two valence electrons result in two series of atomic energy levels as the electron spins can be parallel (triplet states) or anti parallel (singlet state). The energy diagram can be simplified as shown in Fig. 1. The ground state is \(^1S_0\), and the lowest excited states \(nsnp\) are \(^1P_1\) and \(^3P_J\) which can be divided into three fine structure sublevels \(^3P_2, ^3P_1, ^3P_0\). For the \(^3P_J\) state, transitions to higher states can be divided into three groups: \(^3P_J, ^3P_J, ^3P_J\) and \(^3P_J, ^3P_J, ^3P_J\).

By second-order perturbation theory, the energy shift \(U(\omega, p, m)\) of atomic state \(|i\rangle\) with energy \(E_i\) and Zeeman sublevel \(m_i\), which is induced by a trapping laser field with frequency \(\nu=\omega/2\pi\), polarization \(p\), and irradiance intensity \(|I|\) can be expressed as \(U(\omega, p, m)=-a_i(\omega, p, m)I/2\epsilon_0c\) with the

\[\begin{align*}
ns(n+1)P_0 & \equiv ns^1S_0 \\
(nsnp)3P_1 & \equiv ns^1P_1 \\
3P_0 & \equiv 3P_1 \\
3P_2 & \equiv 3P_2 \\
s(n-1)d^3D_{1,2,3} & \equiv ns^1S_0 \\
\end{align*}\]
induced polarizability $\alpha_i$.

The polarizability can be calculated by summing up the contributions from all dipole interactions between the fine structure state $|i\rangle$ and $|k\rangle$ with the Einstein coefficient $A_{jk}$ (spontaneous emission rate for $E_i > E_j$), Zeeman sublevels $m_i, m_k$ and transition frequency $v_{jk} = \omega_{jk}/2\pi$.[13][33].

$$\alpha_i = 6\pi c^3 e_0^2 \sum_{k,m,k} A_{jk}(2J_k + 1) \left( \frac{J_j}{m_i} \right)^2 \left( \frac{J_k}{-m_k} \right)^2$$

(1)

where

$$A_{jk} = \frac{e^2}{4\pi c^3} \frac{3h c^5}{2J_i + 1} \left| \langle \beta_i J_i || D || \beta_j J_j \rangle \right|^2$$

(2)

Here $e$ is the electron charge, $h\omega_{jk}$ is the energy difference between fine structure states $|k\rangle$ and $|i\rangle$, $\beta$ denotes other quantum numbers of the state, and $\langle \beta_i J_i || D || \beta_j J_j \rangle$ is the dipole reduced matrix element. The expression in large parentheses in Eq.(1) denotes a $3J$ symbol which describes the selection rules and relative strength of the transition depending on the involved angular momenta $J$, the projection $m$, and the polarization $p$.

If we know $\omega_{jk}$ and $A_{jk}$ in Eq.(1), we can get the polarizability $\alpha_i$. However, typically the literature gives the total transition rate $A_T$ from a given excited state to the fine structure manifold states below. So we need establish the relation

| UpperState | $5\times17\times5$ | $5\times5p$ | $5\times5p^1$ | $5\times5p^2$ |
|------------|------------------|-------------|-------------|-------------|
| 5s$^2$5p$^1$P$_1$ | 21698.48 | 190.01 | 100.00 | 28565.959 |
| 5s$^2$5p$^1$P$_1$ | 34098.44 | 1.87 | 100.00 | 26965.837 |
| 5s$^2$5p$^1$P$_1$ | 38906.90 | 5.32 | 100.00 | 25181.552 |
| 5s$^2$5p$^1$P$_1$ | 41172.15 | 14.9 | 100.00 | 28565.959 |
| 4s$^2$5p$^1$P$_1$ | 41184.47 | 47.1 | 100.00 | 28176.337 |
| 5s$^2$5p$^1$P$_1$ | 42462.36 | 11.6 | 100.00 | 25181.552 |
| 5s$^2$5p$^1$P$_1$ | 43327.94 | 7.6 | 100.00 | 28565.959 |
| 5s$^2$5p$^1$P$_1$ | 45938.26 | 4.88 | 100.00 | 28565.959 |

TABLE I: Sr element: Transition Wavenumbers (WN)(cm$^{-1}$) corresponding to $\omega_{jk}$, Einstein Coefficients for fine structure states $A_T(\times 10^6 s^{-1})$ and Total $A_T(\times 10^6 s^{-1})$ for fine structure states manifold, Correction Factors $\zeta$. The Wavenumber data originate from [24].

| UpperState | $5\times5p^1p_0^1$ | $5\times5p^1p_1^1$ | $5\times5p^2p_2^1$ |
|------------|------------------|-------------|-------------|
| 5s$^2$5p$^1$P$_0$ | 14721.27 | 10.236 | 10.828 | 14534.444 | 29.526 | 1.0421 | 28565.959 |
| 5s$^2$5p$^1$P$_1$ | 23107.193 | 1.0517 | 4.050 | 22920.362 | 4.106 | 1.0230 | 25181.552 |
| 5s$^2$5p$^1$P$_1$ | 26435.920 | 0.954 | 1.0450 | 26257.089 | 2.803 | 1.0216 | 27552.637 |
| 5s$^2$5p$^1$P$_1$ | 28133.680 | 0.525 | 1.0422 | 27946.849 | 1.543 | 1.0216 | 27552.637 |
| 5s$^2$5p$^1$P$_1$ | 29110.080 | 0.320 | 1.0408 | 28923.249 | 0.943 | 1.0208 | 28565.959 |
| 5s$^2$5p$^1$P$_1$ | 34008.26 | 4.88 | 1.0000 | 34038.26 | 4.88 | 1.0000 | 28565.959 |

Here $\zeta$ is the correction factor. The Wavenumber data originate from [24].
between $A_{Tki}$ and $A_{ki}$. We know $A_{Tki}$ can be expressed as:

$$A_{Tki} = e^2 \frac{4 \omega_{ki}^3}{4 \pi \epsilon_0} \frac{1}{2L_k + 1} |\langle \beta_i L_k \parallel D \parallel \beta_i L_i \rangle|^2$$

(3)

Here $\hbar \omega_{Tki}$ is the energy difference between two fine structure manifold states $|k\rangle$ and $|i\rangle$. Using the formula:

$$\langle \beta_k L_k \parallel D \parallel \beta_i L_i \rangle = (-1)^{J_k + s_i + J_i + 1} \delta_{s_k s_i} \frac{1}{(2J_k + 1)(2J_i + 1)}$$

$$\times \sum \frac{J_k}{L_k} \frac{1}{J_i} \frac{1}{S_i} \frac{1}{L_i} \langle \beta_k L_k \parallel D \parallel \beta_i L_i \rangle$$

(4)

and combining Eq.(2) and (3), we can get

$$A_{Jki} = A_{Tki} \times \zeta(\omega_{ki}) \tilde{R}_{ki}$$

(5)

Here

$$\zeta(\omega_{ki}) = \omega_{Jki}^3 / \omega_{Tki}^3$$

(6)

is the energy dependent correction [19], reflecting the alteration on the transition rate due to the effects such as the orbit-spin interaction and the spin-spin interaction which causes the fine structure splitting. And

$$\tilde{R}_{ki} = (2L_k + 1)(2J_i + 1) \times \left( \frac{J_k}{L_k} \frac{1}{J_i} \frac{1}{S_i} \frac{1}{L_i} \right)^2$$

(7)

gives the fraction of the coupling strength between an excited state $|k\rangle$ and a lower state $|i\rangle$. Since the total transition rate $A_{Tki}$ is usually available in the literature, this geometric ratio tells
us how to scale the interaction for a particular fine structure state of interest.

To calculate the wavelength dependent polarizability, we combine Eq.(1) with Eq.(5), and use the known transition frequencies and spontaneous emission rates in the literature. This light polarizability is very sensitive to the Einstein coefficient. However, theoretical and experimental values of magic wavelengths for the optical clock transition obtained in the past can be used to confirm our calculation.

In this paper, we use this method to calculate the light shift for the terahertz clock transition from \(^3P_0\) to \(^3P_1\), \(m = 0\) levels, and from \(^3P_1, m = 0\) to \(^3P_2, m = 0\) levels for boson isotopes with the nuclear spin \(I = 0\). After calculating magic wavelengths for Sr and Ca optical clock transitions and comparing them to the experimental values, we calculate the polarizability of terahertz transition with data collection mainly from Ref. [20, 25–32].

### III. CALCULATION OF MAGIC WAVELENGTH

#### A. Strontium

Using the method above, for Strontium, we first calculate the magic wavelengths of two optical lattice clock transitions with the data listed in Table I and compare the results with experimental values. Then, we calculate the crossing points for terahertz clock transitions where the difference of polarizability is zero. Table I shows Transition Wavenumbers, Einstein Coefficients and Correction Factors for the 5s5p \(^3P_0\), 5s5p \(^3P_1\), and 5s5p \(^3P_2\) states for Sr element. For Einstein Coefficient \(A_{ki}\), first we choose the available updated experimental values in Ref. [26, 28, 31], then we use updated theoretical data in Ref. [27, 30], and for the rest we mainly use theoretical values in Ref. [25].

According to our calculation, the crossing point for the \(^1S_0\) to \(^3P_0\) transition occurs at 813.1 nm, while the crossing point for the \(^1S_0\) to \(^3P_1(m_f = \pm 1)\) transition with linear polarized light takes place at 915.4 nm. Both of those results are in agreement with the experimental values of 813.428(1) nm [24–26] and 914(1) nm [27]. This confirms our calculation procedure.

Fig. 2 and Fig. 3 display the wavelength dependence of the atomic polarizability difference \(\Delta \alpha\) for Sr with trapping laser wavelength around 400 nm and 1650 nm, respectively. The result is scaled by a factor of \(1/(4\pi\epsilon_0\alpha_0^2)\) and the polarizability is given in atomic unit. In Fig.2, for linear polarized light, \(\Delta \alpha\) between \(^3P_1\) and \(^3P_0\) and \(\Delta \alpha\) between \(^3P_2\) and \(^3P_1\) are given in solid and dash dotted lines, respectively. In Fig.3, \(\Delta \alpha\) between \(^3P_1\) and \(^3P_0\) for linear polarized light and \(\Delta \alpha\) between \(^3P_2\) and \(^3P_1\) for circular polarized light are presented. The cross markers are the crossing points where \(\Delta \alpha\) is zero. From Fig.2 and 3, we can know that the magic wavelength for \(^3P_0\) to \(^3P_1, m = 0\) with linear polarized light are 381.2 nm, 413.6 nm, 419.3 nm, 1714 nm and 3336 nm, while for \(^3P_1, m = 0\) to \(^3P_2, m = 0\) are 384.5 nm, 441.9 nm and 511.0 nm. On the other hand, for \(m = 0\) and circular polarization of light, the magic wavelengths for \(^3P_0\) to \(^3P_1, m = 0\) transition are 511.8 nm and 662.8 nm, while for \(^3P_1, m = 0\) to \(^3P_2, m = 0\), the magic wavelengths are 717.7 nm and 1591 nm.

#### B. Calcium

We calculate the polarizabilities using the data in Table II with the same method. Table II shows Transition Wavenumbers, Einstein Coefficients and Correction Factors for the 4s4p \(^1S_0\), 4s4p \(^3P_0\), 4s4p \(^3P_1\) and 4s4p \(^3P_2\) states for Ca. For Einstein Coefficient, we use the updated theoretical values according to Ref. [33], and others are from the data listed in Ref. [32]. In order to check the accuracy of our calculation and the data used, we get the magic wavelength 799.2 nm for the \(^1S_0, m = 0\) to \(^3P_1, m = 0\) optical transition with circularly polarized trapping light, which agrees well with the experimental value \(800.8(22)\) nm [33].

The wavelength dependence of the atomic polarizability difference \(\Delta \alpha\) around 350 nm and 1350 nm are shown with atomic unit in Fig.4 and 5, respectively. The crossing points where the \(\Delta \alpha\) are zero are marked by cross. The magic wavelengths for linear polarization occur at 1361 nm and 2066 nm for clock transition \(^3P_0\) to \(^3P_1\), while for the transition between
level $^3P_1, m = 0$ and $^3P_2, m = 0$ at 312.2 nm, 316.2 nm, 325.4 nm, 344.0 nm and 393.4 nm.

The laser polarization have no effect on the polarizability for the ground state ($J = 0$) because the ac Stark shift is identical with any polarizations. It is also true for $^3P_0$ state. However, the influence of circular polarized laser light is worth study for other states. For $m = 0$, we can obtain the magic wavelengths for the $^3P_0$ to $^3P_1$ clock transition at 301.0 nm and 310.0 nm, while for $^3P_1$ to $^3P_2$ one finds 1318 nm and 2254 nm.

### C. Magnesium

With the completion of the NIST database, the atomic polarizability of the Mg triplet states in the presence of linear and circular polarized light can also be calculated. Table III presents Transition Wavenumbers, Einstein Coefficients and Correction Factors for the $3s^2\,^1S_0, \, 3s3p\,^3P_0, \, 3s3p\,^3P_1$ and $3s3p\,^3P_2$ states for Mg element. Using the data presented in Table III, the magic wavelengths of $^3P_0$ to $^3P_1, m = 0$ transition with linear polarization are 335.6 nm and 399.5 nm. The magic wavelengths are 308.6 nm, 336.5 nm, 406.1 nm for the transition between $^3P_1, m = 0$ and $^3P_2, m = 0$.

For circular polarization of light, the magic wavelengths for the transition $^3P_0$ to $^3P_1, m = 0$ are 307.7 nm, 336.4 nm, 407.8 nm. However, we can not find any magic wavelength for circular laser between level $^3P_1, m = 0$ and $^3P_2, m = 0$.

![FIG. 4: (Color online) Wavelength dependence of the difference between excited state and ground state atomic polarizability around 350 nm for Ca element.](image-url)

For Mg atoms, several optical transitions between the energy levels of terahertz clock transition states and other levels exist, such as 456.5 nm ($3s\,^1S_0 - 3s3p\,^3P_0$), 383.6 nm ($3s3p\,^3P - 3s3d\,^3D$), 309.6 nm ($3s3p\,^3P - 3s4d\,^3D$), 333.2 nm ($3s3p\,^3P - 3s5s\,^3S$) and 517.4 nm ($3s3p\,^3P - 3s4s\,^3S$). Hence, not all the magic wavelengths are good enough for clock transition, because the slope of the light shift difference with the wavelength is too large (shown in the final table IV).
To some extent, a possible magic wavelength near 400 nm is shown in Fig. 6 with the atomic unit. In Fig. 6, $\Delta \alpha$ for $^1P_1 \rightarrow ^3P_0$ transition and $^3P_2 \rightarrow ^1P_1$ transition with different polarization are given. The cross markers reflect the crossing points where the atomic polarizability difference is zero.

**IV. DISCUSSIONS AND CONCLUSIONS**

In summary, we have calculated magic wavelengths for terahertz clock transitions for alkaline-earth atoms. The calculation results are presented in Table IV along with the slopes of the difference of polarizabilities at corresponding magic wavelengths. Depending on the calculation and the current laser development, we recommend 1714nm and 1591nm for Sr terahertz clock, 1361nm and 1318nm for Ca terahertz clock, 399.5nm and 407.8nm for Mg terahertz clock, because the difference of polarizabilities have small slopes at these magic wavelengths, where we ignore the effect of highly excited states and continuum states which can only make little contribution to the wavelength dependent polarizabilities at terahertz region.

In this paper, we are only focusing on the study of possible magic wavelengths of trapping laser for these terahertz clock transitions of Sr, Ca and Mg atoms. These terahertz clock transitions were first proposed as early as 1972 [15], and recently have been proposed to be applied in active optical clock [34]. These clock transitions of alkaline-earth atoms correspond to a 0.6 THz to 11.8 THz frequency region. After the successful developments of microwave fountain frequency standards, optical clocks with trapped ions and optical lattice trapped neutral atoms, it is interesting to study clock transitions at terahertz wavelengths. The advantages and disadvantages of terahertz magic atomic clock will be discussed elsewhere. The wavelength range studied in this paper (from 500 µm to 25 μm) corresponding to THz frequency standards fills the gap between microwaves and optical waves.

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**TABLE IV: Magic wavelengths for terahertz region. L1 is the linear laser for the $^1P_1$ to $^3P_0$ clock transition while C1 is for the circular laser, L2 is the linear laser for the $^3P_1$ to $^3P_2$ clock transition while C2 is for the circular laser. $\kappa$ is the slope of the shift difference of two level of clock transition levels at the corresponding trapping laser wavelength with unit $Hz/nm$, the sign denotes the direction of the change with the shift for the high level minus the lower level. The data are given in the reasonable experiment condition with input power 150 mW focused to a waist of 65 µm as the light intensity 1.1301 $\times 10^8 W/cm^2$.**

|      | $\lambda$ (nm) | $\kappa$ (Hz/nm) | $\lambda$ (nm) | $\kappa$ (Hz/nm) | $\lambda$ (nm) | $\kappa$ (Hz/nm) |
|------|----------------|------------------|----------------|------------------|----------------|------------------|
| $^3P_{0,1,2}$ |                |                  |                |                  |                |                  |
| L1   | 335.6          | -1125            | 399.5          | -103.5           | 381.2          | -615.7           |
|      | 399.5          | 3066             | 54.94          | 413.6            | -322           |
|      | 381.2          | 35879            | 5759           | 441.9            | -314377        |
|      | 1714           | -5.590           |                |                  |                |                  |
|      | 3336           | 9.122            |                |                  |                |                  |
| L2   | 308.6          | -26574           | 336.5          | 1905             | 384.5          | 1792             |
|      | 336.5          | 3162             | 3879           | 441.9            | 5759           |
|      | 406.1          | 3254             | 1641           | 511              | 1697.63        |
|      | 301.0          | 542.9            |                |                  |                |                  |
|      | 393.4          | 1787             |                |                  |                |                  |
| C1   | 307.7          | -3174            | 301.0          | 8610             | 511.8          | 252.8            |
|      | 336.4          | 469.9            | 10472          | 662.8            | -1068          |
|      | 407.8          | 54.76            |                |                  |                |                  |
| C2   | 1318           | -3.365           | 717.7          | 2692             |                |                  |
|      | 2254           | 13.39            | 5191           | -5.551           |                |                  |

FIG. 5: (Color online) Wavelength dependence of the difference between excited state and ground state atomic polarizability around 1350 nm for Ca element.

FIG. 6: (Color online) The wavelength dependence of the difference of atomic Polarizability around 400 nm for Mg element.
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