Systematic Shifts for Ytterbium-ion Optical Frequency Standards

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The projected systematic uncertainties of single trapped Ytterbium-ion optical frequency standards are estimated for the quadrupole and octupole transitions which are at wavelengths 435.5 nm and 467 nm, respectively. Finite temperature of the ion and its interaction with the external fields introduce drift in the measured frequency compared to its absolute value. Frequency shifts due to electric quadrupole moment, induced polarization and excess micromotion of the ion depend on electric fields, which are estimated in this article. Geometry of the trap electrodes also result in unwanted electric fields which have been considered in our calculation. Magnetic field induced shift and Stark shifts due to electro-magnetic radiation at a surrounding temperature are also estimated. At CSIR-NPL, we are developing a frequency standard based on the octupole transition for which the systematic uncertainties are an order of magnitude smaller than that using the quadrupole transition, as described here.

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I. INTRODUCTION

Recent advances in trapping and laser control of single ion has started a new era for frequency standards [1] in optical frequency region, which can achieve 2-3 orders higher accuracy and lower systematic uncertainty [2] than current microwave clocks based on Cesium fountains [3, 4]. Realization of more accurate frequency standards will open up possibilities of vastly higher speed communication systems and more accurate satellite navigation systems besides enabling more precise verification of fundamental physical theories, in particular related to general relativity [5], cosmology [6], and unification of the fundamental interactions [7]. So far a number of different ion species have been studied as promising optical frequency standards at several research institutes worldwide. These are $^{195}$Hg$^+$ at NIST, USA [5]; $^{171}$Yb$^+$ at NPL, UK [9] & PTB, Germany [10, 11]; $^{115}$In$^+$ at MPQ, Germany [12]; $^{88}$Sr$^+$ at NRC, Canada [13] & NPL, UK [14]; $^{40}$Ca$^+$ at CAS, China [15] & NICT, Japan [16] and $^{27}$Al$^+$ at NIST, USA [2]. The accuracy of a frequency standard is decided by that of the measured atomic transition frequency, which may shift due to inter-species collisions and their interactions with external fields. Therefore, it is important to determine these systematic shifts precisely in order to improve the accuracy of the realized frequency standard.

At CSIR-NPL, India we are presently developing an optical frequency standards using Ytterbium-ion [17]. It has a narrow $^2S_{1/2}; F = 0, m_F = 0$-$^2D_{3/2}; F = 2, m_F = 0$ quadrupole transition (E2) and an ultra-narrow $^2S_{1/2}; F = 0, m_F = 0$-$^2F_{7/2}; F = 3, m_F = 0$ octupole transition (E3) at wavelengths 435.5 nm and 467 nm, respectively [18]. These E2 and E3-transitions are at frequencies $\nu_o = 688 \, 358 \, 979 \, 309 \, 306.62$ Hz [10] and 642 \, 121 \, 496 \, 772 \, 645.15$ Hz [11] with natural line-widths 3.02 Hz and 1 nHz, respectively. We shall be probing the E3-transition in our frequency standards. The nuclear spin $I = 1/2$ of $^{171}$Yb$^+$ allows to eliminate the first-order Zeeman shift. The states associated with the E3-transition have the highest sensitivity to measure temporal constancy of fine structure constant and electron-to-proton mass ratio [19]. In this article we have estimated five major sources of systematic uncertainties which are due to the electric quadrupole shift, Doppler shift, dc Stark shift, black-body radiation shift and Zeeman shift.

II. TRAPPING OF THE YTTERBIUM-ION

A Paul trap [20] of end cap geometry [21] as shown in Fig.1(a) will be employed for trapping single $^{171}$Yb$^+$ ion [17]. For a pure harmonic trapping potential $\Phi^{(k=2)}(x,y,z)$ the time dependent trajectory of the ions [22] can be approximated as

$$u(t) = C \cos \left( \beta_u \frac{\omega_{rf} t}{2} \right) \left[ 1 - \frac{q_u}{2} \cos(\omega_{rf} t) \right]$$

where $u \in \{x,y,z\}$, $C$ is the amplitude of the motion, $\omega_{rf}$ is the applied rf, $\beta_u = \sqrt{a_u + q_u^2/2}$, for $a_u$ and $q_u \ll 1$. The stability parameters, $a_u$ and $q_u$ depend on the applied dc and ac voltages, respectively. For precision measurements, in a real trap the anharmonic trapping potential $\Phi^{(k>2)}(x,y,z)$ [17] are non-negligible. Only the even order multipoles contribute in the case of a cylindrically symmetric end cap trap and the dominating perturbation arise from the octupole term $\Phi^{(k=3)}(x,y,z)$. Neglecting the asymmetries, which may arise from misalignment of the electrodes and machining inaccuracies, the trapping
FIG. 1: (a) The electrode assembly of our end-cap type Paul trap, where \( 2z_0 \approx 0.6 \) mm, \( 2z_2 \approx 1.0 \) mm, \( 2r_1 = 1 \) mm, \( 2r_2 = 1.4 \) mm, \( 2r_3 = 2 \) mm, \( \Theta_1 = 10^\circ \) and \( \Theta_0 = 45^\circ \). (b), (c) and (d) show axial trap depth with respect to radio-frequency \( \omega_{r,f} \), ac \( V \) and dc \( U \) voltages respectively.

The potential can be written as,

\[
\Phi(x, y, z) = \frac{V_T(t)}{2R^2} \left[ c_2(2z^2 - x^2 - y^2) - \frac{c_4}{R^2}(3x^4 + 3y^4) + 8z^4 - 24x^2z^2 - 24y^2z^2 + 6x^2y^2 \right]
\]

where, \( R = \sqrt{r_0^2/2 + z_0^2} \), \( V_T(t) = U + V \cos(\omega_{r,f}t) \) in terms of the dc component \( U \), ac component \( V \) of the trapping voltage and the dimensionless coefficients \( c_2 \), \( c_4 \) depend on electrode geometry. We have simulated geometry dependent trap potential using a commercial software and characterized its nature for several trap geometries as given in Eq. (2). For the trap geometry shown in Fig. 1 the coefficients \( c_2 \) and \( c_4 \) have been estimated to be 0.93 and 0.11, respectively. The restoring force for trapping ions due to \( \Phi(k=2) \) produces an axial trap depth, \( D_z(U, V, \omega_{r,f}) = U/2 + mz^2 \omega_{r,f}^2 q_z^2/16Q \) where \( Q \) and \( m \) are charge and mass of the ion, respectively. Figure 1 (b-d) shows variation of the axial trap depth, \( D_z \) as a function of the control parameters \( U, V \) and \( \omega_{r,f} \). The stability region \( q_z = -16QVc_2/m\omega_{r,f}^2 \) and \( a_z = 32QUc_2/m\omega_{r,f}^2 \) lie in the stability region. Throughout this article we have considered radial coordinate \( r \) in the \( xy \)-plane instead of \( x, y \) coordinates, since the trap is axially symmetric.

III. ELECTRIC QUADRUPOLE SHIFT

Electric quadrupole shift \( \Delta \nu_Q \) of the atomic energy levels is one of the dominating systematic uncertainties for the precision frequency measurement. It arises due to the interaction of the atomic quadrupole moment \( \Theta(\gamma, J) \) of a state having spectroscopic notation \( \gamma \) and total angular momentum quantum number \( J \) with the external electric field gradient \( \nabla E \), giving a Hamiltonian as

\[
H_Q = \nabla E \cdot \Theta = \sum_{q=-2}^{2} (-1)^q \nabla E_q \Theta_{-q}.
\]

The quadrupole moment operator \( \Theta \) and electric field gradient \( \nabla E \) are tensors of rank two \[24\]. A non-zero atomic angular momentum results in a non-spherical charge distribution and the atom acquires a quadrupole moment. The ground state \([S_1/2; 0, 0]\) of \(^{171}\text{Yb}^+\) has \( \Theta(S, 1/2) = 0 \), but the excited states \([2D_{3/2}; 2, 0]\) and \([2F_{7/2}; 3, 0]\) contribute to \( \Delta \nu_Q \). The expectation value of \( H_Q \) in reduced form, as given in Ref. \[25\], is

\[
\langle \gamma J FM_F | H_Q | \gamma J FM_F \rangle = \Theta(\gamma, J) F_Q(1, J, F, m_F)
\]

where \( D_{0q} \) are rotation matrix elements for projecting components of \( \nabla E \) from the principle axes frame that is defined by the trap axes to the lab frame which is defined by the quantization direction \[26\] and

\[
F_Q = (-1)^{\gamma+J+2}(2F+1) \begin{pmatrix} F & 2 & F \\ -m_F & 0 & m_F \end{pmatrix}^{-1} \begin{pmatrix} J & 2 & J \\ -J & 0 & J \end{pmatrix} \begin{pmatrix} F & I & F \end{pmatrix}.
\]

Here the quantities within \( (), \{ \} \) are 3j, 6j-coefficients, respectively and \( F \) is total angular momentum with its projection along the quantization axes \( m_F \). The calculated \( F_Q \) for both \([2D_{3/2}; 2, 0]\) and \([2F_{7/2}; 3, 0]\) states is 1. Due to axial symmetry of the trap the contributions from \( D_{0\pm1} \) cancel with each other and \( D_{00} = (3\cos^2 \theta - 1)/2 \). \( D_{0\pm2} = \sqrt{3/8} \sin^2 \theta(\cos 2\phi \mp i \sin 2\phi) \) contribute to Eq. 4 where \( \Theta \) and \( \phi \) are Euler’s angles that rotates the principle axes frame and overlaps with the lab frame. The tensor components of \( \nabla E \) can be calculated from \( E_{x,y,z} \) produced by \( \Phi(x, y, z) \) as described in Ref. \[24\], which gives for harmonic and anharmonic potentials, respectively.

The measured values of \( \Theta(\gamma, J) \) for the \([2D_{3/2}; 2, 0]\) and \([2F_{7/2}; 3, 0]\) states of \(^{171}\text{Yb}^+\) are 2.08/(11) \( e_a^2 \) \[27\] and \(-0.041(5) e_a^2 \) \[11\] respectively, where \( e \) is electronic charge and \( a_o \) is Bohr radius.

The harmonic component of the trapping potential gives a constant electric field gradient however a spatial dependence comes from the anharmonic component, which introduces an uncertainty in the measured \( \Delta \nu_Q \) due to motion of the ion. We estimate the quadrupole shift due to \( U \) since the contribution from the rf averages to zero for first order electric quadrupole shift and for second order it is zero in case of \(^{171}\text{Yb}^+\). Figure 2 shows the estimated fractional quadrupole shifts \( \Delta \nu_Q/V_0 \) due to \( \Phi^{(2)} \) and \( \Phi^{(4)} \) for the E2 and E3 - transitions of \(^{171}\text{Yb}^+\).
\[
\frac{\Delta \nu_{Q}}{\nu_{o}} = \frac{v^{2}}{2c^{2}} = \frac{2\pi k}{mc^{2}}
\]

for kinetic energy \(\varepsilon_{k}\) of the ion; \(c\) is speed of light. For a laser cooled ion at 1 mK the fractional frequency uncertainty due to the temperature dependent second order Doppler effect is \(\approx 10^{-19}\).

Velocity of the trapped ion can be calculated from its trajectory, which gets deviated from Eq. (1) due to slowly varying stray electric fields. This can result from the patches of unwanted atoms on the electrode surface and relative phase differences of the rf on them. Over the time, Tantalum electrodes get coated with \(^{171}\)Yb atoms coming out of the oven. The differential work-function of Ytterbium and the Tantalum results in an electric field \(E_{p}\), which varies slowly with the deposition of atoms. As a result of an extra force, \(Q\vec{E}_{p}\), the minimum of the confining potential shifts by an amount \(C_{o} = Q\vec{E}_{p} \cdot \vec{u}/(m \omega_{c}^{2})\) and the micromotion increases [32]. A difference in path lengths and non-identical dimensions of the electrodes introduce a phase difference \(\phi_{o}\) between the rf on the electrodes as \(V \cos(\omega_{rf} \pm \phi_{o}/2)\). For small \(\phi_{o}\), i.e. \(\sin\phi_{o} = \phi_{o}/2\), one can approximate this as two parallel plates separated by \(2z_{o}/a\) and at potentials...
\[ U = 0 \text{V} \quad \nu = 500\text{V} \quad \nu_{0} = 2\pi 12 \text{MHz} \]

\[ |\Delta \nu_{D2}| / \nu_{0} \]

\[ U_{\nu} = 500\text{V} \quad \omega_{rf} = 2\pi \times 12 \text{MHz} \]

\[ x10^{-19} \quad x10^{-11} \quad x10^{-14} \]

\[ T (\text{mK}), E_{p}[\text{V/mm}], \phi_{o}[\text{degree}] \]

\[ T [\text{mK}], E_{p}[\text{V/mm}], \phi_{o}[\text{degree}] \]

\[ B_{2} \]

\[ o \]

\[ 2 \]
where $B$ is magnetic field, $\alpha$ is emissivity of the material and $\omega$ is frequency of EM-radiation. The wavelength corresponding to the maximum of the spectral energy density at 300 K is 9.7 $\mu$m [39], which is large compared to the longest transition wavelength $\approx 2.4$ $\mu$m in $^{171}$Yb$^+$. Therefore to a good approximation the BBR generated RMS amplitude of $E$ and $B$ fields are written as $\langle E^2 \rangle = E_o^2 \times (T/300)^4$ and $\langle B^2 \rangle = B_o^2 \times (T/300)^4$, where $E_o = 831.9$ V/m and $B_o = 2.775 \times 10^{-6}$ T, respectively [40].

The magnetic field contributes to a Zeeman shift, which will be discussed in the section VI. The contribution due to $\alpha_2$ can be neglected for an isotropic EM-radiation and the effective BBR shift can be written as

$$\Delta \nu_{BBR} = -\frac{1}{2\hbar} \Delta \alpha_0 E_o^2 \left( \frac{T}{300} \right)^4.$$  \hspace{1cm} (13)

At room temperature a shift of about 0.36 Hz and 0.068 Hz are estimated for the E2 and E3-transitions, respectively.

VI. ZEEMAN SHIFT

Zeeman shift arises due to the interaction of atomic and nuclear magnetic moments $\mu_J$ and $\mu_I$ with an external magnetic field. In an experiment, magnetic field appears from the BBR, geomagnetic and stray fields. The E2 and E3-clock transitions are insensitive to the linear Zeeman effect since ground and excited states have $m_F = 0$ states associated with them. Since the nuclear $g$-factor $g_I$ is much smaller than the electronic $g$-factor $g_J$, the second order Zeeman shift [41] of the sublevels can be approximated only in terms of $g_I$ as

$$\Delta \nu_{QZ} = -\left( \frac{g_J e B}{4\pi m} \right)^2 \sum_{F'} \frac{|F_Z(I, J, F, F', m_F)|^2}{\Delta \nu_{HFS}},$$ \hspace{1cm} (14)

where $\Delta \nu_{HFS}$ is the hyperfine splitting of the states and the matrix element $F_Z = \langle F', m'_F | J_z | F, m_F \rangle$ [42] is given as

$$F_Z = \sqrt{I(I+1)(2I+1)(2F+1)(2F'+1)} \left( \begin{array}{ccc} F & F' & I \\ -m_F & 0 & 1 \end{array} \right).$$ \hspace{1cm} (15)

The calculated $|F_Z|^2 = 1/4$ for the $^2S_{1/2}$, $^2D_{3/2}$ and $^2F_{7/2}$ states in $^{171}$Yb$^+$. Their $g_I$ values are 1.998, 0.8021, 1.1429 and $\Delta \nu_{QZ}$ are 12.643 GHz, 0.86 GHz, 3.62 GHz, respectively [43]. The geomagnetic field in New Delhi, India is approximately 50 $\mu$T which produces $\Delta \nu_{QZ}$ of 38.75 Hz, 91.15 Hz, and 44.19 Hz at the $^2S_{1/2}$, $^2D_{3/2}$ and $^2F_{7/2}$ states, respectively. This results in a net second order Zeeman shift of 52.40 Hz and 5.44 Hz for the E2 and E3-clock transition, respectively. These are much larger than the shift produced by the magnetic field of the BBR at the room temperature whose values are 0.16 Hz and 0.017 Hz for the E2 and E3-transitions, respectively.

VII. CONCLUSION

| Systematic effect | E2-transition $\times 10^{-17}$ | E3-transition $\times 10^{-18}$ |
|-------------------|-------------------------------|-------------------------------|
| Electric quadrupole | 762                          | -789                          |
| Second order Doppler | -1                           | -1                            |
| dc Stark           | -0.09                        | -0.07                         |
| BBR: dc Stark      | -52.3                        | -106                          |
| Second order Zeeman | -3.04                        | -3.39                         |
| BBR: second order Zeeman | -23.2                       | -26.4                         |

The systematic shifts from different source have been estimated for the E2 and E3-transitions of $^{171}$Yb$^+$ and summarized in Tab. I. Even though the electric quadrupole shift is the largest, averaging the measured frequency along three orthogonal directions effectively cancels $\Delta \nu_{QZ}$. Three pairs of Helmholtz coils will be installed for defining the quantization axes. These coils will be used to cancel the static stray magnetic fields as well, for minimizing the quadratic Zeeman shift. The thermal part of $\Delta \nu_{D2}(T)$ is an order of magnitude smaller compared to the frequency standard that we aim for. Careful wiring for supplying rf and accurate machining of the electrodes is very important for making $\phi_0 \approx 0^\circ$. Two pairs of electrodes will be installed in the radial plane for compensating the local electric fields that a trapped ion feels, which is required for minimizing $\Delta \nu_{D2}(E_p)$ and...
\[ \Delta \theta_{dc} \] Surrounding temperature at the position of ion needs to be measured accurately for estimating the Stark and Zeeman shifts produced by BBR, which is the dominating systematic effect (Tab. I). From our estimation, the E3-transition can provide an order of magnitude accurate frequency standards than the E2-transition of \(^{171}\text{Yb}^+\).

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