Evaluation of the performance of a $^{40}\text{Ca}^+ - ^{27}\text{Al}^+$ optical clock

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We report the evaluation of the systematic shifts and the total uncertainty of an $^{27}\text{Al}^+$ optical clock sympathetically cooled by a $^{40}\text{Ca}^+$ ion. Due to its relatively low Doppler cooling limit and the suitability of its mass ratio to the $^{27}\text{Al}^+$, the time dilation shift of the secular motion of our $^{40}\text{Ca}^+ - ^{27}\text{Al}^+$ clock is smaller than the shift of a $^{25}\text{Mg}^+ - ^{27}\text{Al}^+$ clock. The total systematic uncertainty of our $^{40}\text{Ca}^+ - ^{27}\text{Al}^+$ quantum logic clock has been estimated to be $7.6 \times 10^{-18}$, which was mainly limited by the uncertainty of the quadratic Zeeman shift.

INTRODUCTION

Optical clocks have been considered as promising candidates for the next definition of the International System of Units (SI) second [1,2]. A wide range of applications in the study of fundamental physics [3] such as searching for dark matter [4,5] or testing general relativity [6,7], have been proposed. Single-ion optical clocks based on $^{27}\text{Al}^+$ have reached a fractional frequency uncertainty as low as $9.4 \times 10^{-19}$ [8].

Unlike other single-ion optical clocks, $^{27}\text{Al}^+$ single-ion optical clocks depend on another kind of co-trapped ions to provide sympathetic cooling [9] and quantum logic readout [10] because the wavelength of the cooling laser of $^{27}\text{Al}^+$ ion is 167 nm, which is a big challenge until now. Different ion species, such as $^{25}\text{Mg}^+$ [11], $^{40}\text{Ca}^+$ [12] and $^{9}\text{Be}^+$ [13,14] could be employed as the auxiliary ions. However the differences between these auxiliary ions are not obvious.

For an $^{27}\text{Al}^+$ optical clock operating at the Doppler cooling limit, uncertainties from time dilation shift due to the motion is a dominant contribution to the total clock uncertainty [11]. Research has shown that using a $^{40}\text{Ca}^+$ ion as the coolant ion, it is possible to achieve a lower Doppler cooling energy not only because $^{40}\text{Ca}^+$ has a relatively narrower Doppler cooling transition, but also because its mass ratio to $^{27}\text{Al}^+$ is appropriate [15].

In this paper, we report our recent experimental progress in the evaluation of an $^{27}\text{Al}^+$ single-ion optical clock sympathetically cooled by a $^{40}\text{Ca}^+$ ion. Our results show that the time dilation shift due to secular motion of our clock is nearly equal to half of the time dilation shift of a clock sympathetically cooled by $^{25}\text{Mg}^+$ [11], which agrees with the theoretical expectation. We also describe the clock system and present the evolution of the total systematic shifts and uncertainties of the clock transition.

EXPERIMENTAL SETUP

We used a linear Paul trap that is similar to that described previously [16-18]. The electrodes were made of beryllium copper to reduce the coupling capacitor. The distance between the electrodes to the ion was narrowed to 0.4 mm. A pair of end-cap electrodes were placed 8.5 mm apart along the trap axis (z-axis). With an RF drive frequency of $\nu_{RF} = 42.9$ MHz, we achieved a trap frequency of $(\omega_x , \omega_y , \omega_z) = (4.1, 4.1, 1.1)$ MHz.

An Nd: YAG laser at 1064 nm with a maximum pulse energy of 15 µJ and a pulse duration of 2 ns is used for the ablation loading of both $^{40}\text{Ca}^+$ and $^{27}\text{Al}^+$ ions from two separated metal targets. Laser cooling of the $^{40}\text{Ca}^+$ ion is implemented by a 397 nm beam that is approximately...
10 MHz detuned from resonance. Another 397 nm pre-cooling beam with 120 MHz detuning is co-aligned with the Doppler cooling beam to help the ions recrystallize after collision with the background gas. A circularly polarized 397 nm laser beam (SP beam) is applied along the magnetic field direction to initialize the $^{40}\text{Ca}^+$ ion to the state $^2\text{S}_{1/2}(m = -1/2)$.

In the opposite direction, a 267 nm laser not only initialize $^{27}\text{Al}^+$ to either the state $^1\text{S}_0(m = +5/2)$ or $^1\text{S}_0(m = -5/2)$ by switching between two different polarization states, but also helps to perform the quantum logic spectroscopy, with a 729 nm laser beam along the trap axis (729-axial beam). Together with two other 729 nm beams (729-horizontal and 729-vertical), we can detect the excess micromotion in three directions.

Before each $^{27}\text{Al}^+$ clock interrogation pulse, the ions are pre-cooled for 1 ms. Then a Doppler cooling pulse of 1 ms is applied to cool the ions close to their Doppler-cooling limit. All Doppler cooling lasers are opened on during the clock interrogation pulse to ensure that the temperature of the ions remains low. A sequence of pulses then maps $^1\text{S}_0$ state of $^{27}\text{Al}^+$ ion to the dark $^2\text{D}_{5/2}(m = -1/2)$ state on $^{40}\text{Ca}^+$ through their shared motional sidebands. The readout process is repeated up to 10 times to reduce the measurement error. As described in our previous paper[10], we normally operated the clock with an interrogation time of 25 ms, corresponding to a Fourier-limited linewidth of 32 Hz.

**ESTIMATION OF CLOCK TRANSITION SHIFTS**

The time dilation shift due to the motion of the $^{27}\text{Al}^+$ ion has contributed to most of the uncertainty among all published works on the $^{27}\text{Al}^+$ ion optical clock. There are two types of motion for trapped ions: micromotion that is driven by the trap RF field at 42.9 MHz and harmonic-oscillator (secular) motion at the ion’s normal frequencies. In both cases, as the ion moves inside an electric field, the total frequency shift needs to include a frequency-dependent term that corresponds to the Stark effect[11]:

$$\frac{\Delta v}{v} = -\frac{E_p}{mc^2} \left(1 + \frac{f}{400\text{MHz}}\right)^2$$

where $E_p$ and $f$ are the energy and frequency of this motion respectively, and $m$ is the mass of the $^{27}\text{Al}^+$ ion.

The secular motion energy is limited mainly by laser cooling. As we the cooling lasers are maintained on during the clock interrogation, the ions will stay close to the sympathetic Doppler cooling limit. Consider a clock ion with mass $m_2 = m$ cooled by another ion with mass $m_1$, the secular motion energy of the clock ion can be written as:

$$E_{\text{SM},i} = \zeta_i \omega_i^2 b_i^2 z_i^2 (\pi_i + 1/2) = \text{TDS}_i (\pi_i + 1/2)$$

where TDS$_i$ stand for the calculated value representing the time dilation shift per quantum number. $z_i = b_i z_{0,i}$ is the mode amplitude of this motion at the ground state, $\omega_i$ is the mode frequency, $z_{0,i} = \sqrt{\hbar/(2m_2\omega_i)}$, $b_i$ is the component of the normalized eigenvector for the mode[15], and $\pi_i$ is the average motional quantum number that can be measured by comparing the amplitude of the red and blue sidebands:

$$\pi_i = \frac{P_{r,i}}{P_{b,i} - P_{r,i}}$$

$\zeta_i$ is a factor that describes a motion driven by the trapping RF field. This motion has a frequency exactly the same as that of the driven field and exists even in an ideal Paul trap. It is called intrinsic micromotion (IMM) to be distinguished from the excess micromotion (EMM), which arises from the imperfection of the trapping potential, or phase shifts between the trap electrodes. The energy of the IMM is approximately the same as the secular energy in the transverse direction for a single ion. For two co-trapped ions, $\zeta_i$ can be expressed as[15]:

$$\zeta_{i,\text{COM}} = 1 + \frac{2\epsilon^2/\mu}{2\epsilon^2/\mu - 2\alpha - (1 - \sqrt{\mu b_1/b_2})}$$  \hspace{1cm} (4)

$$\zeta_{i,\text{STR}} = 1 + \frac{2\epsilon^2/\mu}{2\epsilon^2/\mu - 2\alpha - (1 + \sqrt{\mu b_2/b_1})}$$  \hspace{1cm} (5)

COM and STR indicate for center-of-mass mode and stretch mode, respectively. $\mu = m_2/m_1$, $\alpha$ and $\epsilon$ are geometric parameters for the trapping field.

One of our measurent is shown in Table IWe take several measurements in different days and take the weighted average of as the final result $-6.9(\pm 3.1) \times 10^{-18}$. The uncertainty is given by twice the standard deviation of the measurements and is shown in the gray band in Fig. 2 (a). Our clock was operated close to the Doppler cooling limit. The secular motion energy at this cooling limit can be calculated[15]:

$$E_{i,\text{limit}} = \frac{\hbar \Gamma}{24} \left(1 + 3l_i^2\right) l_i^2$$

where $\Gamma = 20.4$ MHz is the natural linewidth of the $^2\text{S}_{1/2} \rightarrow ^2\text{P}_{3/2}$ transition on $^{40}\text{Ca}^+$ that is used for the Doppler cooling. $l_i$ represent the projection of the cooling laser to $i$-th direction. The calculated cooling limited is listed as $\pi_c$ in Table I. This corresponding to a time dilation shift of $6.3 \times 10^{-18}$, smaller than a similar $^{27}\text{Al}^+$ clock that symmetrically cooled by $^{25}\text{Mg}^+$[11].

In addition to the IMM, the ion may suffer from additional kinetic energy arising from the imperfection of the trapping potential. This energy $E_{\text{EMM}}$ can be measured.
TABLE I. One of the measurement results for the time-dilation shift due to secular motion. $z$ stands for the amplitude of the motion at the ground state. $\pi_m$ represent the measured average motional quantum number. $\pi_c$ is the calculated average motional quantum number at Doppler cooling limit. TDS/quantum is the calculated time-dilation shift per quantum numbers that has included the contribution of intrinsic micromotion. For this particular measurement, the time-dilation shift is $8.34 \times 10^{-18}$. We made several measurements on different days and take the average as our final result.

| Mode     | $\hat{z}$-COM | $\hat{x}$-STR | $\hat{y}$-COM | $\hat{y}$-STR | $\hat{z}$-COM | $\hat{z}$-STR |
|----------|----------------|---------------|---------------|---------------|---------------|---------------|
| Frequency(MHz) | 4.00           | 2.84          | 4.03          | 2.84          | 1.17          | 2.10          |
| $z$(nm)   | 6.6            | 6.6           | 6.4           | 6.4           | 0.5           | 7.1           |
| $\pi_m$   | 5.2            | 7.0           | 3.0           | 5.7           | 12.8          | 7.5           |
| $\pi_c$   | 3.0            | 4.6           | 3.0           | 4.6           | 7.3           | 4.0           |
| TDS/quantum($10^{-18}$) | 0.747          | 0.004         | 0.757         | 0.003         | 0.030         | 0.118         |
| Total TDS $10^{-18}$ | 3.94           | 0.03          | 2.29          | 0.02          | 0.39          | 0.89          |

Through the ratio of the Rabi rate of the carrier and the EMM sideband $\eta$ [19]:

$$E_{EMM} = \sum_{i} \frac{n_i \nu_{RF}^2 \eta_i^2}{k_L^2 c^2}$$

(7)
i is summed over all three perpendicular directions, and $k_L$ is the wave vector of the detection laser beam.

Our measurement of $E_{EMM}$ is performed on the $^{40}$Ca$^+$ ion ($m_{Ca} = m_1$) instead of the $^{27}$Al$^+$ ion ($m_{Al} = m_2$).

Moreover, only have the vertical and axial beams are perpendicular to each other, while the horizontal beam is at 45° angle to the axial beam (Fig. 1). Taking this into account, the shift on $^{27}$Al$^+$ ion can be written as:

$$E_{EMM} = \sum_{i} \frac{n_i \nu_{RF}^2 \eta_i^2}{m_2 \nu_1^2} (\chi_i \eta_i)^2$$

(8)

where $\nu_1 \approx 411.042$ THz represents the frequency of the detection laser that excited the $^2S_{1/2} \rightarrow ^2D_{5/2}$ transition of the $^{40}$Ca$^+$ ion. $\chi_i$ denotes a factor that describes the projection of laser direction.

During clock operation, we started from minimizing the EMM shift by adjusting the compensation voltages, and left it free-running for the rest of the day. Therefore we continued measuring the EMM shift throughout the entire day and found the total frequency shift due to EMM by averaging those data, yielding a result of $-(3.9 \pm 2.9) \times 10^{-18}$. The uncertainty is given as twice the standard deviation, as shown in the gray band in Fig. 2 (c).

In a perfect linear Paul trap, EMM does not exist along the trap axis. However our trap is not perfect as there is a minimum axial EMM at one spatial location at a spatial location (Fig. 2 (d)). It’s hard to keep the $^{27}$Al$^+$ ion stays at this point because random background-gas collision switched the order of our $^{40}$Ca$^+$ – $^{27}$Al$^+$ pair approximately every 1000 s. We did not control the order of our ions; instead, we pushed the center of our ion pair close to this minimum point and left it free running throughout the entire day. Our measurement includes this shift due to reordering events.

The first-order Doppler shift cannot be observed for most optical clocks that work in the Lamb-Dicke regime because the motional amplitude is much smaller than the laser wavelength. However, a first-order Doppler shift may still occur when the ion itself moves in a fashion correlated with the clock laser. This movement may come from the contraction of the structure of the ion trap or additional electric field generated by the UV photodetection effect.

We monitored this slow but long-term directional
motion by tracking the position of the ions using an Electron-Multiplying charged-coupled device (EMCCD). Our EMCCD was positioned perpendicular to the horizontal plane (the plane of the clock laser), and the motion was detected in the laser plane. The distance between the two ions is 5.3 pixels, which can be used to calibrated the distance between two ions, as this distance is calculated to be 4.9 µm at \( \omega_z = 1.12 \text{ MHz} \). The mean velocity can be calculated based on this.

The motion of ions on the EMCCD is periodic, with the same period as the temperature change period caused by the air conditioner in our laboratory. This is because the EMCCD is supported on a high platform and its supporting structure is susceptible to temperature. To eliminate this influence, we performed a moving average for one hour of data to calculate the mean velocity, as shown in Fig. 3 (a) and (b). \(^{40}\text{Ca}^+ - ^{27}\text{Al}^+ \) sometimes change their order along the trap axis due to the collision of the background gas with the ion pair. Therefore we took the center position of the Z-direction of the ion pair to eliminate this effect.

The velocity of ions varies within ±0.1 nm/s. We assumed that the upper limit of the ion motion speed is 0.1 nm/s, which may still be caused by the EMCCD’s support structure changes affected by temperature. This leads to a first-order Doppler shift of \((0 \pm 0.2) \times 10^{-18} \). Although the ion trap was in an ultra-high vacuum chamber, collisions from background gas still exist causing secular motion heating and clock phase shifts. The evaluation of the frequency shift resulting from collisions depends on the vacuum pressure \( p \). The energy \( e \) is required by the \(^{40}\text{Ca}^+ - ^{27}\text{Al}^+ \) ions to change their order is given by \(^{21}\):

\[
E_{\text{reorder}} = \frac{3}{4} \left( \frac{\sqrt{m_0 e^2}}{2\pi e_0} \right)^{2/3} \times \left( \frac{2(e^2 + \alpha - 1)(e^2 + \mu(\alpha - 1))}{e^2(\mu + 1) + 2\mu(\alpha - 1)} \right)^{1/3} - 1 \tag{9}
\]

where \( e \) is the charge of the electron. The relationship between the vacuum pressure and the reorder rate is given by:

\[
\Gamma_{\text{reorder}} \lesssim \frac{1}{2} \left( \frac{p}{902 \text{nPa}} \right) \left( \frac{E_{\text{reorder}}}{1 \text{K} \times k_B} \right) \tag{10}
\]

In our \(^{27}\text{Al}^+ \) clock, the reorder event can be easily observed on the EMCCD camera\(^{21}\)) because only the position of the \(^{40}\text{Ca}^+ \) ion, which is the bright spot, can be seen. We measured the image for a period of 0.5 s and averaged 10 times to reduce noise. This averaging normally does not miss any reorder events as the reorder period is around 1000 s.

The reorder rate is given by \( \Gamma_{\text{reorder}} = N_{\text{reorder}}/T \), where \( N_{\text{reorder}} \) is the number of reorder events during time \( T \). In our case, the reorder rate was measured to be \( \Gamma_{\text{reorder}} = 0.0013 \text{ s}^{-1} \), corresponding to a vacuum pressure of 3.8 nPa. Using the empirical formula from Rosenband et. al.\(^{13}\), and assuming that each collision brings a maximum phase shift of \( \pi/2 \), the frequency shift due to background gas collision is estimated to be 0.195 mHz, corresponding to a fractional frequency shift of 0.2 \( \times 10^{-18} \).

External magnetic influence is the dominant effect that causes a frequency shift of the clock transition in our \(^{27}\text{Al}^+ \) clock. Considering the first-order and second-order terms in the magnetic field, the two atomic resonance frequencies will be shifted \(^{21}\):

\[
\nu = \nu_0 + C_1 B + C_2 (B^2) \tag{11}
\]

where \( \nu_0 \) is the unperturbed resonance frequency and \( C_1, C_2 \) are the coefficient quantifying the linear and the quadratic Zeeman shift, respectively. Here, the linear Zeeman shift is compensated by the interleaved locking of the magnetic field measured in real time, and \( B_{ac}^2 \) is an oscillating (ac) magnetic field driven by the power line or the RF field.

\[
\langle B \rangle = 3.664 \times 10^{-4} \text{ T} \text{ can be measured with high accuracy by interleaved locking of the transitions } ^2S_{1/2}(m = -1/2) \rightarrow ^2D_{5/2}(m = -5/2) \text{ and } ^2S_{1/2}(m = +1/2) \rightarrow ^2D_{5/2}(m = +5/2) \text{ in the } ^{40}\text{Ca}^+ \text{ ion. For the ac component } \langle B_{ac} \rangle, \text{ we only observe frequency jitter under 150 Hz, which results in a coherence time of 81.6 µs. This}
\]
corresponds to $B_{ac} = B_{rms} = \langle B \rangle \pm 1.795 \times 10^{-7}$ T. With $C_2 = -7.9144(24) \times 10^7 \text{Hz/T}^2$, the fractional frequency shift due to the quadratic Zeeman effect is $-(8621.0 \pm 6.2) \times 10^{-18}$.

The blackbody radiation (BBR) leads to an AC Stark shift in the clock transition. Our $^{27}\text{Al}^+$ clock is operated at room temperature ($\approx 294$ K), and the BBR temperature is mainly caused by the thermal radiation emitted from various components of the ion trap. We built another ion trap system with exact same configuration to achieve an accurate assessment of the BBR temperature using an infrared thermal imager. The finite element analysis method was also employed to achieve an accurate assessment of the BBR temperature[22]. The temperatures of the various components in the vacuum chamber are listed in Table II.

For the $^{27}\text{Al}^+$ ion, the effective BBR temperature can be calculated using:

$$T_{eff} = \sqrt{\sum_i \frac{\Omega_{eff} T_i^4}{4\pi}}$$

(12)

where $T_{eff}$ is the BBR temperature felt by the ion, and $\Omega_{eff}$ and $T_i$ are the effective solid angle and temperature of each component of the trapped ion system, respectively. Considering the difference between the simulation and with the actual system, the uncertainty will increase slightly. Based on a conservative estimate, for the ion trap system evaluated above, we evaluated the BBR temperature as $T_{eff} = 299.6 \pm 1.4$ K. So that the clock frequency shift due to BBR is[23]:

$$\Delta \mu = -\pi k I T_{eff}^4 (\Delta \alpha_0 \times 1.00024)$$

(13)

where $\Delta \alpha_0 = 4\pi \epsilon_0 \times (1.03\pm0.11) \times 10^{-31} \text{m}^3$ is the static differential polarizability. The corresponding BBR shift is evaluated as $-(5.3 \pm 0.6) \times 10^{-18}$.

The electric field of the optical radiation incident on the ion perturbs and shifts the line center of the ion transition. This contributes to the Stark shift[19]:

$$\Delta v_{ac} = -\frac{\alpha_{ac}(\lambda)}{2\epsilon_0} I$$

(14)

TABLE II. BBR temperature evaluation results of the ion trap system

| Components                  | $\Omega_{eff}$ | Value (K) | Uncertainty (K) | Uncertainty $T_{eff}$ (K) |
|-----------------------------|---------------|-----------|-----------------|--------------------------|
| Blade electrode             | 0.457         | 303.87    | 2.90            | 0.110                    |
| Cap electrode               | 0.293         | 304.16    | 2.59            | 0.064                    |
| Insulation support          | 2.284         | 302.78    | 2.58            | 0.488                    |
| Compensation electrode      | 0.245         | 304.07    | 2.65            | 0.055                    |
| Stainless steel bracket     | 0.675         | 302.78    | 2.21            | 0.128                    |
| Chamber                     | 1.264         | 297.97    | 1.43            | 0.169                    |
| Glass Windows               | 6.016         | 297.97    | 1.43            | 0.801                    |
| Flange                      | 1.333         | 297.97    | 1.43            | 0.178                    |

TABLE III. Fractional frequency shifts and uncertainties for the $^{27}\text{Al}^+$ quantum-logic clock

| Effect                        | Shift($10^{-18}$) | Uncertainty($10^{-18}$) |
|-------------------------------|-------------------|--------------------------|
| Quad.Zeeman                   | -8621.0           | 6.2                      |
| Secular motion                | -6.9              | 3.1                      |
| Excess micromotion            | -3.9              | 2.9                      |
| Blackbody radiation           | -5.3              | 0.6                      |
| Cooling laser Stark           | -12.3             | 0.3                      |
| AOM freq. error               | 0                 | 0.3                      |
| First-order Doppler           | 0                 | 0.2                      |
| Background-gas collision      | 0                 | 0.2                      |
| clock laser Stark             | 0                 | 0.1                      |
| Total                         | -8627.7           | 7.6                      |

where $\Delta \alpha_0(\lambda)$ is the dynamic polarizability of the wavelength $\lambda$ and I is the intensity of the incident light. The intensity of the clock laser was measured to be 40 nW, focused on an area of 120 µm in diameter, corresponding to an AC Stark shift $(0 \pm 0.1) \times 10^{-18}$.

In addition to the clock laser itself, both the 397 nm cooling light and the 866 nm pumping light are present during clock operation. The intensity focus of those lights on the ion was $55 \pm 20$ nW and $91 \pm 4$ µW with a spot diameter of 80 µm and 150 µm, respectively. The AC Stark shifts are $-(0.042 \pm 0.015) \times 10^{-18}$ and $-(12.22 \pm 1.89) \times 10^{-18}$, respectively. The total AC Stark shift caused by the cooling lasers is $-(12.3 \pm 0.3) \times 10^{-18}$.

Phase chirp in the clock beam AOM can also contribute to a frequency shift as the optical path through the crystal changes when it switches on and off. The shift due to a phase chirp is approximately[13] $df \approx 0.2P(1-d)f$ Hz, where $P$ is the RF power drives the AOM and $d$ is duty cycle of the AOM. We drive 1.698 mW AOM RF power to generate a 40 nW clock laser on the ion, so the shift due to this phase chirp in the clock beam AOM is $-(0 \pm 0.3) \times 10^{-18}$.

We have evaluated the fractional frequency shift and associated systematic uncertainties for our $^{27}\text{Al}^+$ clock. The results are shown in the Table[13] and the total shift and associated systematic uncertainty are $-(8627.7 \pm 7.6) \times 10^{-18}$. The systematic uncertainty is limited by the quadratic Zeeman shift, which is mainly caused by the measurement uncertainty of the quadratic Zeeman effect.
coefficient $C_2$, and reducing the magnetic field $\langle B \rangle$ may lead to a significant improvement. Our measurement of the secular motion temperature is also limited by decoherence from magnetic field noise and jitter on the amplitude of the RF trapping field. In addition, controlling the order of the ions is inevitable that the uncertainty of time dilation shift due to excess micromotion can be improved.

**CONCLUSION**

The latest development of an $^{27}$Al$^+$ quantum-logic atomic clock sympathetically cooled by a $^{40}$Ca$^+$ ion has been reported. Owing to the relatively lower Doppler cooling limit from the $^{40}$Ca$^+$ ion and the suitability mass ratio to $^{27}$Al$^+$ ion, we achieved a smaller time-dilation shift due to secular motion than that of an $^{25}$Mg$^+$ – $^{27}$Al$^+$ clock. We further studied other systematic shifts and that of the other $^{27}$Al$^+$ quantum-logic atomic clock. In conclusion, we have built an $^{27}$Al$^+$ quantum-logic atomic clock with a total fractional frequency shift and associated systematic uncertainty of $-(8627.7 \pm 7.6) \times 10^{-18}$.

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