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SENSITIVITY OF THE H_3O^+ INVERSION–ROTATIONAL SPECTRUM TO CHANGES IN THE ELECTRON-TO-PROTON MASS RATIO

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ABSTRACT

Quantum-mechanical tunneling inversion transition in ammonia (NH_3) is actively used as a sensitive tool to study possible variations of the electron-to-proton mass ratio, $\mu = m_e/m_p$. The molecule H_3O^+ has the inversion barrier significantly lower than that of NH_3. Consequently, its tunneling transition occurs in the far-infrared (FIR) region and mixes with rotational transitions. Several such FIR and submillimeter transitions are observed from the interstellar medium in the Milky Way and in nearby galaxies. We show that the rest-frame frequencies of these transitions are very sensitive to the variation of μ, and that their sensitivity coefficients have different signs. Thus, H_3O^+ can be used as an independent target to test hypothetical changes in μ measured at different ambient conditions of high (terrestrial) and low (interstellar medium) matter densities. The environmental dependence of μ and coupling constants is suggested in a class of chameleon-type scalar field models—candidates to dark energy carrier.

Key words: dark energy – elementary particles – ISM: molecules – molecular data – techniques: radial velocities

1. INTRODUCTION

The spatial and temporal variability of dimensionless physical constants has become a topic of considerable interest in laboratory and astrophysical studies as a test of the Einstein equivalence principle of local position invariance (LPI), which states that outcomes of nongravitational experiments should be independent of their position in spacetime (e.g., Dent 2008). The violation of LPI is anticipated in some extensions of the Standard Model and, in particular, in those dealing with dark energy (e.g., Hui et al. 2009; Damour & Donoghue 2010).

A concept of dark energy with negative pressure ($p = -\rho$) appeared in physics long before the discovery of the accelerating universe through observations of nearby and distant (at redshift $z \sim 1$) Type Ia supernovae (Perlmutter et al. 1998; Riess et al. 1998). Examples of dark energy in a form of a scalar field with a self-interaction potential can be found in reviews by Peebles & Ratra (2003), Copeland et al. (2006), and by Uzan (2010). Since that time many sophisticated models have been suggested to explain the nature of dark energy and among them the scalar fields which are ultra-light in cosmic vacuum but possess a large mass locally when they are coupled to ordinary matter by the so-called chameleon mechanism (Khoury & Weltman 2004; Brax et al. 2004, 2010a, 2010b; Avelino 2008). A subclass of these models considered by Olive & Pospelov (2008) predicts that fundamental physical quantities such as elementary particle masses and low-energy coupling constants may also depend on the local matter density. Since the mass of the electron m_e is proportional to the Higgs vacuum expectation value (VEV $\sim$ 200 GeV), and the mass of the proton m_p is proportional to the quantum chromodynamics (QCD) scale $\Lambda_{QCD} \sim 220$ MeV, we may probe the ratio of the electroweak scale to the strong scale through measurements of the dimensionless mass ratio $\mu = m_e/m_p$ in a high-density laboratory (terrestrial) environment, μ_{lab}, and in low-density interstellar clouds, μ_{space} ($\rho_{lab}/\rho_{space} > 10^{10}$). In this way, we are testing whether the scalar field models have chameleon-type interaction with ordinary matter. Several possibilities for detecting chameleons from astronomical observations were discussed in Burrage et al. (2009), Davis et al. (2009), Brax & Zioutas (2010), and Avgoustidis et al. (2010). First experiments constraining these models were recently carried out in Fermilab (Upadhye et al. 2010) and Lawrence Livermore National Laboratory (Rybka et al. 2010).

At the moment, the most accurate relative changes in the mass ratio $\Delta\mu/\mu = (\mu_{space} - \mu_{lab})/\mu_{lab}$ can be obtained with the ammonia method (van Veldhoven et al. 2004; Flambaum & Kozlov 2007). NH_3 is a molecule whose inversion frequencies are very sensitive to any changes in μ because of the quantum-mechanical tunneling of the N atom through the plane of the H atoms. The sensitivity coefficient to μ-variation of the NH_3 $(J, K) = (1, 1)$ inversion transition at 24 GHz is $Q_{inv} = 4.46$. This means that the inversion frequency scales as $\Delta\omega/\omega = 4.46(\Delta\mu/\mu)$. In other words, sensitivity to μ-variation is 4.46 times higher than that of molecular rotational transitions, where $Q_{rot} = 1$. Thus, by comparing the observed radial velocity of the inversion transition of NH_3, V_{inv}, with a suitable rotational transition, V_{rot}, of another molecule arising co-spatially with ammonia, a limit on the spatial variation of μ can be determined,

$$\frac{\Delta\mu}{\mu} = \frac{V_{rot} - V_{inv}}{c(Q_{inv} - Q_{rot})} \approx 0.3\frac{\Delta V}{c}, \quad (1)$$

where c is the speed of light and $\Delta V = V_{rot} - V_{inv}$.

Surprisingly, recent observations of a sample of nearby (distance $R \sim 140$ pc) cold molecular cores ($T_{kin} \sim 10$ K, $n = 10^4$–10^5 cm^{-3}, $B < 10\,\mu$G) in lines of NH_3 $(J, K) = (1, 1)$ at 24 GHz, HC_3N $J = 2 - 1$ at 18 GHz, and N_2H^+ $J = 1 - 0$ at 93 GHz reveal a statistically significant positive velocity offset between the low-J rotational and inversion transitions, $\Delta V = V_{rot} - V_{inv} = 27 \pm 4_{stat} \pm 3_{sys}$ m s^{-1}, which gives $\Delta\mu/\mu = (26 \pm 4_{stat} \pm 3_{sys}) \times 10^{-9}$ (Levshakov et al. 2010b).[5]

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[5] Presented are the corrected values of ΔV and $\Delta\mu/\mu$ discussed in Levshakov et al. (2010a).

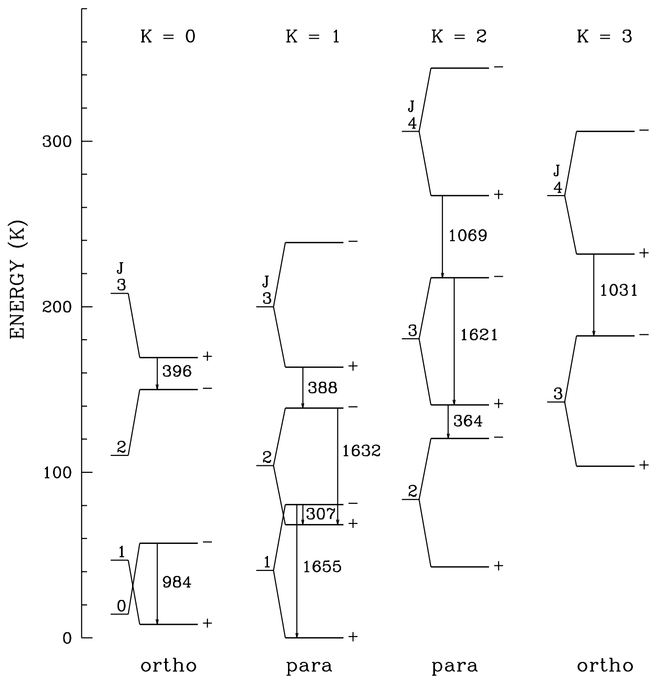

Figure 1. Level scheme for H_3O^+. The depicted frequencies are in GHz.

A few molecular cores from this sample were mapped in the NH_3 (1,1) and HC_3N (2–1) lines, and it was found that in two of them (L1498 and L1512) these lines trace the same material and show the offset $\Delta V = 26.9 \pm 1.2_{\rm stat} \pm 3.0_{\rm sys}$ m s^{-1} throughout the entirety of the clouds (Levshakov et al. 2010a). It was also demonstrated that for these clouds the frequency shifts caused by external electric and magnetic fields and by the cosmic blackbody radiation-induced Stark effect are less than 1 m s^{-1}. Optical depth effects in these clouds were studied from the analysis of unsaturated ($\tau < 1$) and slightly saturated ($\tau \approx 1$–2) hyperfine components of the corresponding molecular transitions, and it was found that both groups of lines have similar radial velocities within the 1σ uncertainty intervals.

The nonzero $\Delta\mu$ implies that at deep interstellar vacuum the electron-to-proton mass ratio increases by $\sim 3 \times 10^{-8}$ as compared with its terrestrial value and, hence, LPI is broken. In view of the potentially important application of this discrepancy to the fundamental physics, one has to be sure that the nonzero $\Delta\mu$ is not caused by some overlooked systematic errors. An obvious way to tackle this problem is to use other molecular transitions, which have different sensitivity coefficients Q_μ. It has already been suggested for measuring Λ-doublet lines of light diatomic molecules OH and CH (Kozlov 2009), and microwave inversion–rotational transitions in partly deuterated ammonia NH_2D and ND_2H (Kozlov et al. 2010).

In this paper, we propose to use tunneling and rotation transitions in the hydronium ion H_3O^+. Like ammonia, it also has a double minimum vibrational potential. The inversion transitions occur when the oxygen atom tunnels through the plane of the hydrogen atoms. This leads to an inversion splitting of the rotational levels. The splitting of H_3O^+ is very large, 55.3462 ± 0.0055 cm^{-1} (Liu & Oka 1985), as compared to the 1.3 cm^{-1} splitting in NH_3. Consequently, the ground-state inversion–rotational spectrum of H_3O^+ is observed in the submillimeter-wave region (Plummer et al. 1985; Bogey et al. 1985; Verhoeve et al. 1988), whereas pure inversion transitions are observed in the far-infrared (FIR) region (Verhoeve et al. 1989; Yu et al. 2009).

H_3O^+ has both ortho- and para-modifications (see Figure 1). In the submillimeter—the range accessible from high-altitude ground-based telescopes—there are three low-lying transitions at 307, 364, and 388 GHz which belong to para-H_3O^+, and one ortho-H_3O^+ transition at 396 GHz. The 388 GHz line is, however, blocked by water vapor in the atmosphere. The other lines were observed in the interstellar molecular clouds (Hollis et al. 1986; Wootten et al. 1986; Wootten et al. 1991; van der Tak et al. 2006; Phillips et al. 1992). The 364 GHz line was also observed in two galaxies: M82 and Arp 220 (van der Tak et al. 2008). In far-IR, H_3O^+ lines were detected from aboard the space observatories at $\omega = 4.31$ THz (Timmermann et al. 1996), $\omega = 1.66$, 2.97, and 2.98 THz (Goicoechea & Cernicharo 2001; Lerate et al. 2006; Polehampton et al. 2007), $\omega = 984$ GHz (Gerin et al. 2010), and $\omega = 1.03$, 1.07, and 1.63 THz (Benz et al. 2010).

The observed transitions of H_3O^+ arise in the warm ($T_{\rm kin} \sim$ 100 K) and dense ($n \approx 10^5$–10^6 cm^{-3}) star-forming regions surrounding protostars, where hydronium appears to be one of the most abundant species with the abundance as high as $X(H_3O^+) \approx 5 \times 10^{-9}$ (Wootten et al. 1991; Benz et al. 2010).

2. SENSITIVITY COEFFICIENT OF INVERSION TRANSITION

Sensitivity of the H_3O^+ inversion transition to μ-variation can be estimated from the analytical Wentzel–Kramers–Brillouin (WKB) approximation. Following Landau & Lifshitz (1977), we write for the inversion frequency (units used are $\hbar = |e| = m_e = 1$)

$$\omega_{\rm inv} \approx \frac{2E_0}{\pi}\, e^{-S}, \tag{2}$$

where S is the action over classically forbidden region and E_0 is the ground-state vibrational energy. Expression (2) gives the following sensitivity to μ-variation:

$$Q_{\rm inv} \approx \frac{S+1}{2} + \frac{S\,E_0}{2(U_{\rm max} - E_0)}\,, \tag{3}$$

where $U_{\rm max}$ is the barrier height and we are not using an additional approximation $E_0 = \omega_v/2$.

According to Rajamäki et al. (2004) and Dong & Nesbitt (2006), we can take $U_{\rm max} = 651$ cm^{-1} and $E_0 \approx 400$ cm^{-1}. The inversion frequency for H_3O^+ is 55.3 cm^{-1}. Thus, Equations (2) and (3) give

$$S \approx 1.5\,, \qquad Q_{\rm inv} \approx 2.5\,. \tag{4}$$

Dong & Nesbitt (2006) report the inversion frequencies for H_3O^+, H_2DO^+, HD_2O^+, and D_3O^+ to be 55.3 cm^{-1}, 40.5 cm^{-1}, 27.0 cm^{-1}, and 15.4 cm^{-1}, respectively. Neglecting the weak dependence of the reduced mass, m_r, on the inversion coordinate, we take m_r to be 0.7, 0.8, 1.0, and 1.25, respectively (Dong & Nesbitt 2006). Figure 2 shows the inversion frequency as a function of m_r. From this plot, we can estimate the sensitivity coefficient for H_3O^+ to be

$$Q_{\rm inv} \approx 2.46\,, \tag{5}$$

which is in perfect agreement with Equation (4). We can conclude that the inversion transition in H_3O^+ is almost two times less sensitive to μ-variation than a similar transition in NH_3, where $Q_{\rm inv} = 4.5$ (Flambaum & Kozlov 2007).

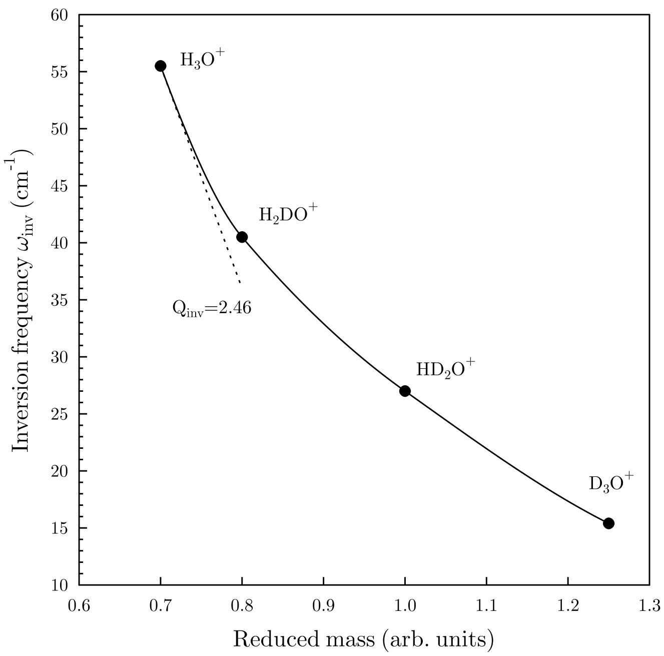

Figure 2. Inversion frequency as a function of the reduced mass for hydronium ion isotopologues.

3. SENSITIVITY COEFFICIENTS OF MIXED TRANSITIONS

The spectrum of the rotational and inversion transitions of H_3O^+ is studied in Yu et al. (2009). For the lowest vibrational state, we can write the simplified inversion–rotational Hamiltonian as

$$H = BJ(J+1) + (C-B)K^2 - D_J[J(J+1)]^2 - D_{JK}J(J+1)K^2 - D_K K^4 + \cdots + \frac{s}{2}\{W_0 + W_J J(J+1) + W_K K^2 + \cdots\}. \quad (6)$$

Here we neglected higher terms of expansion in J and K; $s = \pm 1$ for symmetric and antisymmetric inversion state; total parity $p = (-1)^K s$. Numerical values are given in Yu et al. (2009; MHz): $B = 334{,}406$; $C - B = -148{,}804$; $D_J = 35$; $D_{JK} = -70$; $D_K = 41$; $W_0 = -1659{,}350$; $W_J = 5988$; and $W_K = -8458$. Note that we write Hamiltonian (6) in such a way that terms which determine inversion splitting are collected in the last line. Therefore, we have the following relation with parameters used in Yu et al. (2009):

$$B = [B(0^+) + B(0^-)]/2\,, \quad W_J = B(0^+) - B(0^-)\,, \quad (7)$$

and similarly for $C - B$ and W_K. Parameters D_J, D_{JK}, and D_K are averaged over inversion states $s = \pm 1$.

To estimate sensitivities of the mixed transitions, it is sufficient to account for μ-dependence of the dominant parameters B, C, and W_0. It is clear that B and $C \sim \mu$, and W_0 scales as $\mu^{Q_{inv}}$. It follows that for rotational part of the energy we have $Q_{rot} = 1$ and for inversion part Q_{inv} is given by Equation (4) or (5). This leads to the expressions, used earlier for NH_2D (Kozlov et al. 2010),

$$\omega_{mix} = \omega_{rot} \pm \omega_{inv} \quad (8)$$

and

$$Q_{mix} = \frac{\omega_{rot}}{\omega_{mix}} Q_{rot} \pm \frac{\omega_{inv}}{\omega_{mix}} Q_{inv}\,. \quad (9)$$

Table 1
Frequencies and Sensitivities to μ-Variation of the Inversion–Rotation Transitions in H_3O^+

Transition						Frequency (MHz)			Q_μ
J	K	s	J'	K'	s'	Experiment	Error	Equation (6)	
1	1	−1	2	1	+1	307192.410	0.05	307072	+9.0
3	2	+1	2	2	−1	364797.427	0.10	365046	−5.7
3	1	+1	2	1	−1	388458.641	0.08	389160	−5.2
3	0	+1	2	0	−1	396272.412	0.06	397198	−5.1
0	0	−1	1	0	+1	984711.907	0.30	984690	+3.5
4	3	+1	3	3	−1	1031293.738	0.30	1031664	−1.4
4	2	+1	3	2	−1	1069826.632	0.30	1071154	−1.2
3	2	−1	3	2	+1	1621738.993	2.00	1621326	+2.5
2	1	−1	2	1	+1	1632090.98		1631880	+2.5
1	1	−1	1	1	+1	1655833.910	1.50	1655832	+2.5

Note. Experimental frequencies are taken from Pickett et al. (1998) and Yu et al. (2009).

We use Hamiltonian (6) and Equation (9) to calculate the frequencies and sensitivities of the mixed transitions. The obtained results are presented in Table 1. Final results are very sensitive to the parameter Q_{inv}. A good agreement between two different estimates of Q_{inv} from Equation (4) and Figure 1 shows that this parameter is known with 10% accuracy, or better. In the next approximation, we need to weight independently all terms of the Hamiltonian (6) with different scalings. However, this does not lead to any significant changes in sensitivities, Q_μ, of the low-J transitions from Table 1.

4. DISCUSSION AND CONCLUSIONS

We have shown above that the rest-frame frequencies of the inversion–rotational transitions of H_3O^+ are very sensitive to the value of μ. For a given transition from Table 1, ω_i, with the sensitivity coefficient Q_i, the expected frequency shift, $\Delta\omega_i/\omega_i$, due to a change in μ is given by Levshakov et al. (2010b)

$$\frac{\Delta\omega_i}{\omega_i} \equiv \frac{\tilde{\omega}_i - \omega_i}{\omega_i} = Q_i \frac{\Delta\mu}{\mu}\,, \quad (10)$$

where ω_i and $\tilde{\omega}_i$ are the frequencies corresponding to the laboratory value of μ and to an altered μ in a low-density environment, respectively.

By analogy with Equation (1), we can estimate the value of $\Delta\mu/\mu$ from two transitions with different sensitivity coefficients Q_i and Q_j

$$\frac{\Delta\mu}{\mu} = \frac{V_j - V_i}{c(Q_i - Q_j)}\,, \quad (11)$$

where V_j and V_i are the apparent radial velocities of the corresponding H_3O^+ transitions.

Consider the two lowest frequency transitions from Table 1: $1_1^- \to 2_1^+$ and $3_2^+ \to 2_2^-$ of para-H_3O^+ at 307 and 364 GHz, respectively. Here, $\Delta Q = Q_{307} - Q_{364} = 14.7$, which is four times larger then the ΔQ value from the ammonia method. This means that the offset $\Delta V \sim 27$ m s^{-1}, detected in the ammonia method, should correspond to the relative velocity shift between these transitions, $\Delta V = V_{364} - V_{307}$, of about 100 m s^{-1}.

Published results on interstellar H_3O^+ allow us to put an upper limit on $\Delta\mu/\mu$. The observations of the 307, 364, and 396 GHz lines carried out at the 10.4 m telescope of the Caltech Submillimeter Observatory (CSO) by Phillips et al. (1992) and the observations of the 307 and 364 GHz lines at the 12 m

APEX telescope (Atacama Pathfinder Experiment) by van der Tak et al. (2006) have accuracy of about 1 km s^{-1} that provides a limit on $\Delta\mu/\mu < 2 \times 10^{-7}$, which is consistent with the signal $\sim 3 \times 10^{-8}$ revealed by the ammonia method (Levshakov et al. 2010a, 2010b).

In order to check ammonia results, we need to improve the accuracy of H_3O^+ observations by more than one order of magnitude. According to Table 1, the uncertainties of the laboratory frequencies of the transitions at 307, 364, and 396 GHz are 50, 80, and 45 m s^{-1}, respectively. Therefore, we also need a factor of a few improvement of the laboratory accuracy to be able to detect reliably an expected signal $\Delta V \sim 100$ m s^{-1} and to check the nonzero ammonia results.

An important advantage of the hydronium method is that it is based on only one molecule. In the ammonia method, there is unavoidable Doppler noise caused by relative velocity shifts due to spatial segregation of NH_3 and other molecules. When using hydronium, the only source of the Doppler noise may arise from possible kinetic temperature fluctuations within the molecular cloud since the submillimeter H_3O^+ transitions have different upper level energies: $E_u = 80$, 139, and 169 K for the 307, 364, and 396 GHz transitions, respectively. It is also important that two H_3O^+ transitions have similar Q values: $Q_{364} \approx Q_{396}$. This allows us to control the Doppler noise and to measure accurately the relative position of the 307 GHz line.

The analysis of other possible systematic effects for hydronium is mostly similar to what was done in detail for ammonia in Levshakov et al. (2010a). The systematic shifts caused by pressure effects are about a few m s^{-1}, or lower. As mentioned in Section 1, the frequency shifts caused by external electric and magnetic fields and by the cosmic blackbody radiation-induced Stark effect are less or about 1 m s^{-1} for NH_3.

An additional source of systematic for H_3O^+ can come from the unresolved hyperfine structure (HFS) in combination with possible nonthermal HFS populations in the interstellar medium. As noted by Keto & Rybicki (2010), HFS lines reduce the effective optical depth of the molecular rotational transition by spreading the emission out over a wider bandwidth. To our knowledge, the HFS has not been resolved yet for $H_3{}^{16}O^+$ either in laboratory or astronomical measurements. An expected size of the hyperfine splittings can be estimated using analogy with ammonia. For the latter, the main hyperfine splitting is associated with the spin of nitrogen $\boldsymbol{I}_1$. The maximum hyperfine splitting caused by the hydrogenic spin $\boldsymbol{I}$ is about 40 kHz (Ho & Townes 1983). This splitting includes interaction with the nitrogen spin $\sim(\boldsymbol{I}_1 \cdot \boldsymbol{I})$ and with molecular rotation $\sim(\boldsymbol{J} \cdot \boldsymbol{I})$. In hydronium, the oxygen nucleus is spinless and there is only interaction with rotation $\sim(\boldsymbol{J} \cdot \boldsymbol{I})$. Hydronium has a similar electronic structure and close rotational constants to ammonia, so its spin–rotational interaction should be $\lesssim$40 kHz. Thus, we can expect less than 40 m s^{-1} of the hyperfine bandwidth for 300 GHz lines and smaller splittings at higher frequencies. At $T_{\rm kin} \sim 100$ K, a typical kinetic temperature of warm and dense gas in the star-forming regions, the thermal width of the H_3O^+ lines is comparable with this HFS splitting.

For para-H_3O^+, transitions at 307, 388, and 364 GHz have three HFS components each (two transitions with $\Delta F = \Delta J$ are strong, and the remaining one is weak). For ortho-H_3O^+, the 396 GHz transition has nine HFS components (four strong, three weaker, and two weakest).

The difference in the magnitude of the energies of the hyperfine and rotational transitions in molecular spectra in the submillimeter range is considerable: milli-Kelvin for the hyperfine levels and tens of Kelvin between rotational levels. This means that the HFS levels may be populated approximately in statistical equilibrium even if the rotational levels are not (Keto & Rybicki 2010). Therefore, we may suggest that the line centers of the submillimeter H_3O^+ transitions are not affected significantly by relative populations of the hyperfine levels and that the expected velocity shifts are of a few m s^{-1}.

To sum up, the systematic shifts of the line centers caused by possible nonthermal HFS populations and pressure effects are likely not larger than 10 m s^{-1}, which is about 10% of the expected relative shift between the para-H_3O^+ $J_K = 1_1^- \to 2_1^+$ and $3_2^+ \to 2_2^-$ transitions due to μ-variation. A more accurate analysis of the HFS-induced systematics will be possible after the HFS is either measured or calculated theoretically.

Finally, we would like to note that other isotopologues of the hydronium ion must also have large sensitivity coefficients Q_μ. To use them, we need laboratory studies of the low-frequency mixed transitions in the spectra of the partly deuterated hydronium ions H_2DO^+, HD_2O^+, and in D_3O^+.

In the near future, high-precision measurements in the submillimeter and FIR ranges with greatly improved sensitivity will be available with the Atacama Large Millimeter/submillimeter Array (ALMA), the Stratospheric Observatory For Infrared Astronomy (SOFIA), the Cornell Caltech Atacama Telescope (CCAT), and others. Thus, any further advances in exploring $\Delta\mu/\mu$ depend crucially on accurate laboratory measurements ($\Delta\omega/\omega \lesssim 10^{-8}$) of relevant molecular transitions in the submillimeter and FIR ranges where reliable spectroscopic data are still relatively poor.

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