Search of the variation in the proton-to-electron mass ratio using two vibrational transition frequencies of molecular ions

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We propose to monitor the variation in the proton-to-electron mass ratio (m_p/m_e) from the two vibrational transition frequencies of molecular ions.

We consider the following vibrational transition frequencies $f(v')$

for example $^{16}O_2^+$ $X^2\Pi_{1/2}$ $(v,J)=(0,1/2)-(v',1/2)$　　$f(v') = f_v v' - xf v' (v' + 1)$

$Q(v') = [f(v') - f(2v')/2]/f(v') = xf v'/f_v$

(ratio between harmonic and unharmonic vinratioal terms)

$Q(v')$　　proportional to $(m_p/m_e)^{-1/2}$

$^{16}O_2^+$ $Q(4)$ measured with the uncertainty of 10^{-18}

cancel the frequency shifts satisfying $\delta f(v')/f(v') = \delta f(2v')/f(2v')$

(relativistic effects etc.)

Precise measurement of time & frequency

important role for the development of physics beyond standard model

Atomic transition frequencies

attained the accuracy of **10^{-18}**

$^1S_0$-$^3P_0$ ^{87}Sr(Ohmae-san's talk), ^{171}Yb, ^{27}Al$^+$

(should be possible also with ^{115}In$^+$ Ohtsubo-san's poster)

S-F ^{171}Yb$^+$ →sensitive to the variation in the finestructure constant

$(-4.1 \pm 2.5) \times 10^{-18}$/yr

But molecular transition frequencies has never been measured with the uncertainty lower than 10^{-15}

Why precise measurement of molecular transitions is useful?

We can observe phonomenons, which cannot be observed with atomic transitions

(1) **variation in the proton-to-electron mass ratio**

vibration freq. $\propto (m_p/m_e)^{-1/2}$ rotational freq. $\propto (m_p/m_e)^{-1}$

(2) detection of electron EDM (Abe-san's talk)

(3) simmetry violation of chiral molecules

(4) gravity in the micro size

Which molecular transition is useful for precise measurement?

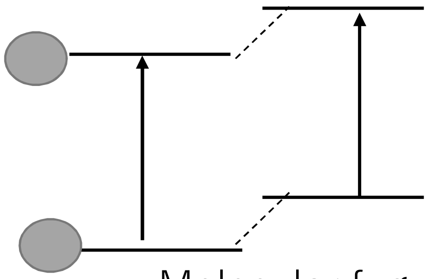
vibrational transition with

$\Delta N = \Delta J = \Delta F = \Delta M = \mathbf{0}$ (only v changes)

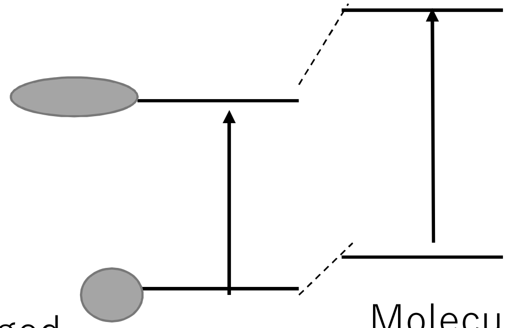
(molecular shape does not change)

↓

Stark, Zeeman, electric quadrupole shifts at upper and lower states are almost equal (cancelled)



Molecular fugure unchanged
Parallel shift,



Molecular shift changes
Shifts are not parallel

Molecular vibrational transition frequency

$$v = 0 \rightarrow \quad v'$$

We can select the convenient transition to prepare the probe laser

Probe laser between **1.3 – 1.5 μm**

linewidth narrower than 10 mHz is possible using cold Si cavity

Natural linewidth of vibratinoal spectrum of diatomic molecules

hetero-nuclear: several Hz

homo-nuclear: < 1 μHz (ultra-narrow laser linewidth is useful)

Why precise measurement of molcular transition is difficult?
How can we overcome with molecular ions (co-trapped with atomic ion)?

Complicated energy structure (vibrational-rotationsl states)

Laser cooling is difficult

sympathetic cooling with laser cooled atomic ions

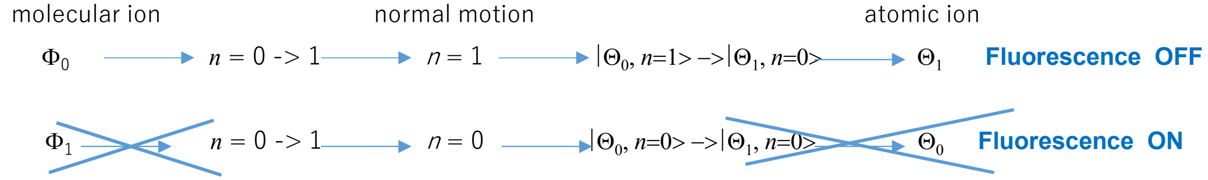
Difficult to localize in a selected state

quantum logical one way transition (sideband transition) n: normal motion mode

repeat $|\Phi_i, n=0\rangle \rightarrow |\Phi_{i+1}, n=1\rangle$ and $|n=1\rangle \rightarrow |n=0\rangle$

Difficult to monitor the state by fluorescence

quantum logical detection

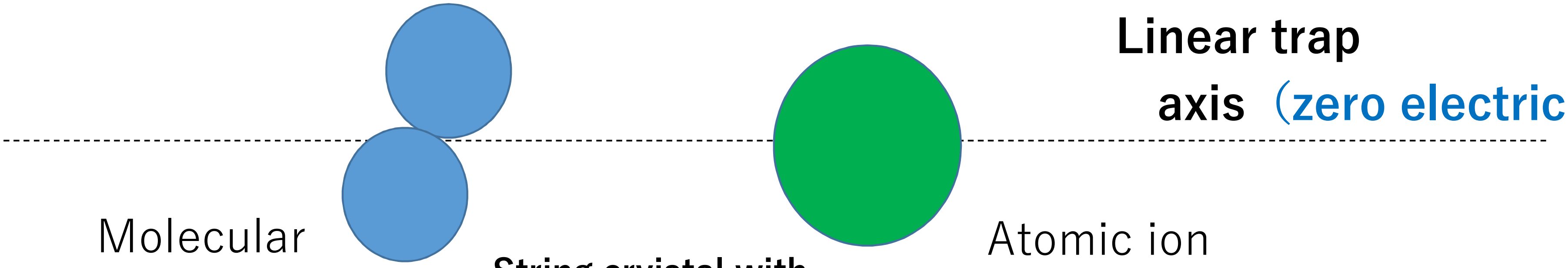


molecular ion   normal motion   atomic ion

$\Phi_0 \rightarrow n = 0 \rightarrow 1 \rightarrow n = 1 \rightarrow |\Theta_0, n=1\rangle \rightarrow |\Theta_1, n=0\rangle \rightarrow \Theta_1$ **Fluorescence OFF**

$\Phi_1 \rightarrow n = 0 \rightarrow 1 \rightarrow n = 0 \rightarrow |\Theta_0, n=0\rangle \rightarrow |\Theta_1, n=0\rangle \rightarrow \Theta_0$ **Fluorescence ON**

Molecular ion in a linear trap
(Sympathetically cooled with atomic ion)



Linear trap axis **（zero electric field）**

Molecular ion

String cryistal with low kinetic energy

Atomic ion
（laser cooled）

For homonuclear molecular ion, there is no electric dipole coupling between different states in the electric ground state

↓

（1） **No measurement perturbation by blackbody radiation**

（2） **Stark is very small**

$^{16}O_2^+$ $X^2\Pi_{1/2}(v,J,M) = (0,1/2,\pm1/2) - (v',1/2,\pm1/2)$ transition frequency

v: vibrational state J: total angular momentum M: component of J parallel to the magnetic field

Electric quadrupole shifts zero

Zeeman shift strict linear with coefficients of 3 x 10^{-15}/G

(much smaller than Al^+clock, Sr and Hg lattice clocks)

Eliminated perfectly averaging $M = \pm 1/2 - \pm 1/2$

DC Stark shift : -3.1 x 10^{-20} /(V/cm)^2

Blackbody radiatin shift (300 K) : -2.0 x 10^{-18}

(same order with Al^+, much smaller than Sr and Hg lattice clocks)

One photon forbidden -> two photon absorption

($v' = 4$: 2.74 μm, $v'=8$; 1.41 μm)

$v' = 4$ light shift = -1.4 x 10^{-14} with 1 Hz Rabi Freq. (800 W/cm^2)

$v' = 8$ light shift = -1.2 x 10^{-14} with 1 Hz Rabi Freq. (800 W/cm^2)

using Hyper Ramsey, supressed to < 10^{-18}

Systematic uncertainty < 10^{-18}

Statistic uncertainty with $^{16}O_2^+$ $(v,J)=(0,1/2)\rightarrow(8,1/2)$

Spectrum linewidth is given by probe laser (**1.41 μm**) linewidth

→ narrower than **10 mHz** can be attained stabilizing with cold Si cavity

Statistic uncertainty assuming

single molecular ion

linewidth of 100 mHz (Rabi freq. **0.1 Hz** and light shift **1.2×10^{-15}**)

statistic uncertainty 6.6×10^{-19} with one day measurement

Attainable accuracy is higher than lattice clocks

Useful to monitor the variation in m_p/m_e using an atomic clock (Sr lattice?) for reference

Can we search without the atomic clock for reference?

Using atomic clock at a distant place, earth tide gives a fluctuation

Search of the variation in m_p/m_e using $Q(v') = [f(v')-f(2v')/2]/f(v')$

$$f(v') = v' f_v - v'(v'+1)\, xf$$

$f_v \propto (m_p/m_e)^{-1/2}$ harmonic term

$xf \propto (m_p/m_e)^{-1}$ unharmmonic term

$$Q(v') = [f(v') - f(2v')/2]/f(v') = xf v'/f_v \propto (m_p/m_e)^{-1/2}$$

Precise measurement of $Q(v')$ is useful for search the variation in (m_p/m_e)

Current upper limit of m_p/m_e 10^{-16} /yr

Frequency shift in $Q(v')$

$$\delta Q(v') = \{ f(2v')/[2f(v') - f(2v')]\} \times [\delta f(v')/f(v') - \delta f(2v')/f(2v')]$$

No shift in $Q(v')$ for the shifts with $\delta f(v')/f(v') = \delta f(2v')/f(2v')$

(1) Quadratic Doppler shift + Gravity red shift   canceled perfectly
(limit of the accuracy of Al^+ clock)

(2) Light shift induced by the probe laser ($\delta f_L(v')$) $\propto$ laser power density $I_p(v')$
the light shift is eliminated optimizing $I_p(v')/I_p(2v')$ so that $\delta f_L(v')/f(v') = \delta f_L(2v')/f(2v')$
if the sign of the light shift is the same for $f(v')$ and $f(2v')$
for O_2^+ transition, $\delta f_L(v')$ is always negative
Hyper Ramsey is useful to eliminate the effect of the fluctuation in $I_p(v')/I_p(2v')$

Other frequency shift in $Q(v')$ with O_2^+ $^2\Pi_{1/2}(v, J) = (0,1/2) \rightarrow (v',1/2)$

Electric quadrupole shift: zero

Zeeman shift: perfect linear with $\pm 1.6 \times 10^{-14}$/G
(eliminated averaging $M = \pm 1/2 \rightarrow \pm 1/2$)

DC Stark: $Q(1)$ $-2.4 \times 10^{-18}/(\mathrm{V/cm})^2$ $Q(2)$ $-2.5 \times 10^{-19}/(\mathrm{V/cm})^2$
$Q(4)$ $8.5 \times 10^{-21}/(\mathrm{V/cm})^2$ (string crystal is not definitely required)

Blackbody radiation shift (300 K): $Q(1)$ -1.5×10^{-16} $Q(2)$ -1.6×10^{-17}
$Q(4)$ 5.4×10^{-19}

Accuracy of 10^{-18} is attainable

We don‘t need an atomic clock for reference

Statistical uncertainty of O_2^+ *Q*(*4*)

Spectrum linewidth is given by the laser linewidth (natural linewidth < 1 μm)

O_2^+ $v = 0 \rightarrow 8$ two photon absorption of **1.41 μm** laser

(linewidth < 10 mHz is attainable using cold Si cavity)

$v = 0 \rightarrow 4$ two photon absorption of **2.74 μm** laser

or

two photon absorption of **signal** and **idler waves** (f_s and f_i)of optical parametric oscillator (OPO) pumped by **1.37 μm**

(pump laser is stabilized within 10 mHz using cold Si cavity)

(no effect with the fluctuation of $f_s \rightarrow f_s + \delta f$, $f_i \rightarrow f_i - \delta f$)

Statistical uncertainty with a single molecular ion with the linewidth of 100 mHz

6×10^{-18} with two weeks

Measurement with multi-molecular ion is also possible

The proposed method is applicable also with other molecular ions satisfying

(1) $\Delta J = 0$ with $J = 0$ or 1/2　(electric quadrupole shift zero)

(2) Transition between stretched states

(Zeeman shift is linear)

(3) Sign of light shift is the same with $f(v')$ and $f(2v')$

(light shift is eliminated by optimizing the intensity ratio of two probe lasers)

Applicable also $(v,J) = (0,0) \rightarrow (v',0)$ **with CaH^+, SrH^+ etc.**

Not applicable with $^{15}N_2^+$ $(v,N,J) = (0,0,1/2) \rightarrow (v',0,1/2)$

(sign of light shift depends on v')

Conclusion

We propose to measure the variation in the proton-to-electron mass ratio (m_p/m_e) using two vibrational transition frequencies of molecular ion

Example: We consider $f(v')$ with

$^{16}O_2^+$ $^2\Pi_{1/2}$ $(v,J) = (0,1/2)$-$(v',1/2)$ electric quadrupole shift zero

Zeemaan shift eliminated perfectly

$$Q(v') = [f(v') - f(2v')/2]/f(v') \propto (m_p/m_e)^{-1/2}$$

elimination of relativistic effects

light shift induced by probe laser

supression of DC Stark shift

blackbody radiation shift

useful for the search of the variation in (m_p/m_e)

atomic clock for reference is not necessary

Publications

M. Kajita, Phys. Rev. A **95**, 023418 (2017)

M. Kajita, J. Phys. Soc. Jpn. **86**, 123301(2017)

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