Assessing the time constancy of the proton-to-electron mass ratio by precision ro-vibrational spectroscopy of a cold molecular beam

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ABSTRACT

We report the design of an experiment that aims to constrain, over a-few-year timescale, the fractional temporal variation of the proton-to-electron mass ratio, $\beta = m_p/m_e$, at a level of 10^{-15}/yr by means of a spectroscopic frequency measurement on a beam of cold CF_3H molecules. This is extracted from a buffer-gas-cooling source and then collimated by means of an electrostatic hexapole lens. Employed in a two-photon Ramsey-fringes interrogation scheme, the probe source is based on a mid-infrared quantum cascade laser, phase-locked to a specially-developed optical frequency comb that is ultimately referenced to the Cs primary standard via an optical fiber link.

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1. Introduction

Thanks to the breakthrough technologies originating respectively from the world of ultracold quantum gases and that of femtosecond laser frequency combs during the last decades, methods of precision spectroscopy have advanced to the point where atomic/molecular transition frequencies can be determined with an astonishing precision (down to a few parts in 10^{-18}), to such an extent that detecting the influence of fundamental new physics at the eV energy scale is now within reach [1]. In this frame, more and more challenging experiments are underway, aiming at testing nature symmetries and constants with unprecedented sensitivity. In particular, in the last few years a strong interest has focused on the possibility that what we know as the fundamental physical constants might show variations over cosmological time scales. Such an effect arises quite naturally in modern theories (Lie Groups, String/M Theories, ...) attempting either to establish a Grand Unification Theory beyond the Standard Model or to reconcile this latter and General Relativity in a Theory of Everything [2–5]. Since variation of dimensional constants cannot be distinguished from that of the units, it makes more sense to consider changes of dimensionless parameters. The prime target is the fine

structure constant, $\alpha = e^2/(4\pi\varepsilon_0\hbar c)$, which defines the scale of quantum electrodynamics; the second prominent quantity is the proton-to-electron mass ratio, $\beta = m_p/m_e$, which characterizes the strength of strong interaction in terms of the electro-weak one. While in the former case the temporal stability is conveniently probed through atomic transitions [6], the β ratio is more accurately addressed with molecular systems where resorting to the inaccurate nuclear Schmidt model is bypassed [7,8]. One approach is to compare the wavelengths of molecular lines measured in the present epoch on Earth with the corresponding ones from astronomical objects at high redshifts [9–11]. The most up-to-date constraint for $\Delta\beta/\beta$ is $(0.0 \pm 1.0) \cdot 10^{-7}$, deduced from the observation of radio-frequency transitions of methanol in the PKS1830-211 galaxy at redshift $z = 0.89$ (corresponding to a look-back time of 7 billion years). Assuming a linear temporal variation, this implies a fractional variation $\dot{\beta}/\beta$ on the order of $10^{-17} \cdot yr^{-1}$, consistent with a null result [12]. A second approach relies on precision molecular spectroscopy experiments entirely carried out in the laboratory, with the inherent advantage that all systematic effects can be meticulously controlled. Different schemes have been proposed in this frame, including an ammonia fountain [13], molecular hydrogen ions confined in radio-frequency traps [14], and magnetically trapped cold XH (X: alkali-earth atom) molecules [15].

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2. The overall proposal

Following the scheme originally implemented by Shelkovnikov and coworkers with a conventional supersonic beam of SF_6 [16], our proposal is to measure the frequency v_{vib} of a given molecular ro-vibrational transition relative to the clock hyperfine transition v_{hyp} in the Cs electronic ground state ($|F = 4, m_F = 0\rangle \leftrightarrow |F = 3, m_F = 0\rangle$). Such frequencies scale respectively as [3]

$$v_{vib}(S) = K_S \cdot \beta^{-1/2} \cdot Ry \tag{1}$$

$$v_{hyp}(Cs) = K_{Cs} \cdot \left(\frac{\mu_{Cs}}{\mu_B}\right) \cdot \alpha^2 \cdot F(\alpha) \cdot Ry \tag{2}$$

where S indicates a generic molecular species. Here, K_S and K_{Cs} are proportionality constants, μ_{Cs} is the magnetic dipole of the Cs nucleus, μ_B the Bohr magneton, Ry the Rydberg constant, and $F(\alpha)$ is a dimensionless function accounting for relativistic effects in Cs, whose dependence on α is to the power of 0.83. From these formulas the following relationship between our observable v_{vib}/v_{hyp} and β is derived

$$\frac{1}{\frac{v_{vib}(S)}{v_{hyp}(Cs)}} \frac{\partial\left[\frac{v_{vib}(S)}{v_{hyp}(Cs)}\right]}{\partial t} = -\frac{1}{2\beta}\frac{\partial\beta}{\partial t} - \frac{2.83}{\alpha}\frac{\partial\alpha}{\partial t} - \frac{1}{\frac{\mu_{Cs}}{\mu_B}}\frac{\partial\left[\frac{\mu_{Cs}}{\mu_B}\right]}{\partial t} \tag{3}$$

which, in combination with the current constraints of $\dot{\alpha}/\alpha = (-1.6 \pm 2.3) \cdot 10^{-17}$ yr^{-1} and $d/dt[\ln(\mu_{Cs}/\mu_B)] = (-1.9 \pm 4.0) \cdot 10^{-16}$ yr^{-1}, as inferred from a 6-year record of increasingly precise atomic clock frequency comparisons [17], can be used to set an upper limit to the temporal variation of β.

To enhance the spectroscopic interrogation time, which sets the ultimate resolution achievable in a single measurement, a Ramsey-fringes technique will be adopted [18]. In this scheme one first wants to minimize the fringe periodicity P, given by the ratio of the mean longitudinal speed of the molecules in the beam, u, to the distance D between the two interaction zones: $P = u/(2D)$. Second, a high fringe contrast is desirable, which can be accomplished by reducing the velocity dispersion, $(\Delta u)^{-1} \sim 1/\sqrt{T_{trans}}$, with T_{trans} being the translational temperature of the molecular beam. As discussed later, a low rotational temperature, T_{rot}, is also desirable; indeed, this increases the population in states with low rotational quantum numbers, which can be exploited to enhance the strength of the final spectroscopic absorption feature. While for a generic molecular beam the inequality chain $T_{vib} \gtrsim T_{rot} \gtrsim T_{trans}$ holds, under typical operating conditions of our experiment (see Section 3.2), these three temperatures roughly coincide with each other so that we will simply speak of the beam temperature T. In conclusion, a great enhancement in the ultimate accuracy can be achieved if a beam of slow and cold molecules is used. In the optical domain, the Ramsey-fringes method has to be associated with a sub-Doppler technique in order to avoid a scrambling of the fringe pattern. Opting for two-photon spectroscopy, the most efficient configuration is represented by the two-zone geometry where the molecular beam interacts successively with two standing waves generated from the probe laser inside a single folded Fabry–Perot enhancement cavity comprising four mirrors in a U configuration [19,20].

In the following, we shall describe the main building blocks of the experiment starting from those which, already constructed, are about to be characterized (Section 3) and ending with those that are being developed (Section 4).

3. Design of the molecular beam

The aim is to generate a molecular beam with intense flux, low temperature, and reduced longitudinal speed. This will be attained by combining buffer-gas cooling (BGC) and Stark manipulation. Since BGC operates with nearly all species, potentially any molecule with a relatively high electric dipole moment (EDM) and a favorable two-photon transition may be chosen for the experiment.

3.1. The molecular choice

The fluoroform (CF_3H) molecule exhibits both these features. First, a fair permanent EDM value, $\mu = 1.67$ Debye, makes it suitable for manipulation with external electric fields. Second, it has a fundamental, strong ro-vibrational band (CF_3) degenerate stretch, v_5) at 8.63 μm (1158.75 cm^{-1}) [21], where high-performance quantum cascade lasers (QCLs) are available and effective optical frequency combs can be developed. Shown in Fig. 1 is the two-photon transition identified for the spectroscopic measurement, $|0\rangle \equiv |v_5 = 0, J = 1, K = 1\rangle \rightarrow |1\rangle \equiv |v_5 = 1, J = 2, K = 2, l = -1\rangle \rightarrow |2\rangle \equiv |v_5 = 2, J = 2, K = 1\rangle$, where J is the total angular momentum quantum number, K describes the projection of the vector $\boldsymbol{J}$ onto the molecular axis and l is an additional quantum number (exclusively for rotational states pertaining to degenerate vibrational levels) from the Coriolis coupling interaction [22]. To determine the detuning δ of the virtual level from the intermediate one, we start with the general consideration that the energy of a given ro-vibrational level is the sum of a vibrational and a rotational term, $E_{ro-vib} = E_{vib} + E_{rot}$. In addition, for an oblate symmetric-top molecule and retaining only the leading terms, the rotational energy can be expressed as $E_{rot} \equiv E(J, K) = BJ(J+1) + (C - B)K^2 - 2\xi Kl$. Then, using the vibrational anharmonicity value
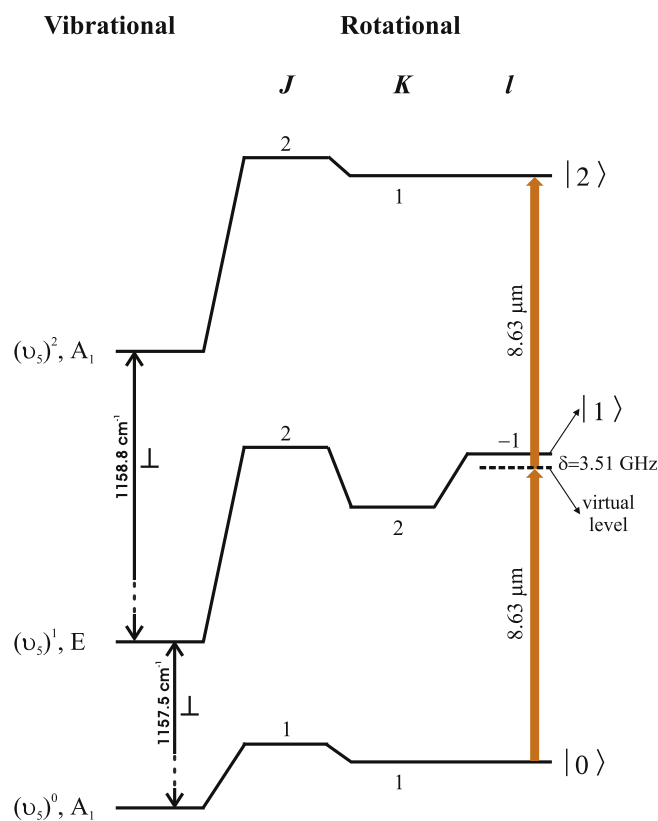
Fig. 1. CF$_3$H two-photon transition selected for the Ramsey interrogation. The symbol $\perp$ indicates that the vibrational transition moment is perpendicular to the top axis; accordingly, the following selection rules hold: $\Delta K \equiv K_i - K_f = \pm 1$; $\Delta J = 0, \pm 1$. In addition, for $(A_1 \rightarrow E)$-type transitions, the following selection rules are valid for the quantum number l: $\Delta l = +1$ for $\Delta K = -1$; $\Delta l = -1$ for $\Delta K = +1$; the reverse is true for $(E \rightarrow A_1)$-type transitions.