

Testing the Time-Invariance of Fundamental Constants Using Cold and Not-So-Cold Molecules

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INTRODUCTION

The equivalence principle of general relativity postulates that the outcome of any non-gravitational experiment is independent of position and time. At a first glance, the equivalence principle seems a bare minimum for a self-consistent theory, but in fact, many theories that attempt to unify gravity with other fundamental forces violate this principle. The Kaluza-Klein theories from the 1920s as well as modern string theories, for instance, introduce additional compactified dimensions, and the size of these – yet unobserved – dimensions determines the strength of the fundamental forces. If the size of these dimensions should happen to change over time, the strength of the forces in four-dimensional space-time would change as well. Such a change would manifest itself as a change of the coupling constants and particle masses [1]. Therefore, by monitoring the time-invariance of these constants one might hope to get a glance of physics beyond the standard model.

With this motivation, many studies have been, and are being, performed to search for variations of fundamental constants. Interestingly, two recent astrophysical studies by Murphy et al. [2] and by Reinhold et al. [3] have reported a small but significant time-variation of α – the fine structure constant representing the strength of the electro-weak force – and the proton-electron mass ratio, $\mu = m_p/m_e$ – which is a measure for the strength of the strong force. Murphy et al. have looked at absorptions of various atoms in the spectra of distant quasars. They make use of the fact that due to relativistic contributions the hyperfine-splitting in atoms depends on $Z\alpha^{-2}$, with Z being the atomic number. By comparing different atomic species they infer both the red shift due to the expansion of the universe (which is used to calculate the look-back time) and α_z/α_0 , where α_z is the fine-structure constant at the epoch when the light was absorbed, and α_0 is the present value of the fine structure constant. Their results suggest that α has changed by 1 part in 100 thousand over the last ten billion years, corresponding to a change of $10^{-15}/\text{yr}$ if one assumes a linear cosmologic expansion model. Reinhold et al. have looked at absorption of molecular hydrogen in quasar systems. The spectra of molecules depend both on α and on the proton-to-electron mass ratio, μ . Reinhold et al. found an indication that μ has changed by two parts in 10^5 over cosmological time. If one assumes that the constants change linearly over time, this implies a fractional change on the order of 10^{-15} per year.

To test the time-variation of fundamental constants in the current epoch, frequency standards based on different atomic and molecular transitions are being compared as a function of time. As these standards have in general a different dependence on α and μ , a possible time-variation of α and/or μ will lead to a frequency shift. The sensitivity of an experiment looking for an effect due to a linear change of a fundamental constant, X , can be expressed as;

$$\left(\frac{\partial X}{X} \right) / \frac{\partial t} = \frac{\partial \nu}{\nu} / \frac{K \cdot \Delta t}{K \cdot \Delta t} \quad (1)$$

with $\left(\frac{\partial X}{X}\right)/\frac{\partial t}{t}$ being the fractional rate of change of X, $\frac{\partial \nu}{\nu}$ being the fractional frequency precision of the

measurement, K being the inherent sensitivity of a transition, defined via $\frac{\partial \nu}{\nu} = K \frac{\partial X}{X}$, and Δt being the time

duration of the experiment. In the hydrogen quasar data the resolution of the recorded spectra is 10^{-6} - 10^{-7} , the sensitivity factor of the observed transitions range from -0.01 to +0.05 and the look-back time is on the order of 10^{10} years. This results in an overall sensitivity (averaged over the 76 observed transitions) for the rate of change of μ on the order of 10^{-15} per year. For obvious reasons, laboratory tests of the time-variation typically span only a few years, however, they have the advantage that one is free to choose a transition in an atom or molecule that allows one to either obtain a very high precision or one that is inherently very sensitive to a variation of a fundamental constants – or preferably both. Currently, the most stringent *independent* laboratory test of the time-variation of μ is set by comparing vibrational transitions in SF_6 with a Cesium fountain over 2 years, which has resulted in a limit for the variation of $\Delta\mu/\mu < 5.6 \times 10^{-14}/\text{yr}$ [4]. We will discuss two experiments aimed at constraining the time-variation of the proto-to-electron mass ratio that are underway in our lab. One experiment uses extreme precision measurements on cold ammonia molecules in a molecular fountain, the other uses the enhanced sensitivity due to near degeneracies between rotational levels in the fine structure ladders of CO in the metastable $a^3\Pi$ state.

A MOLECULAR FOUNTAIN

Ultimately, the precision in any spectroscopic measurement is limited by the interaction time of the particle to be investigated with the radiation field. In conventional molecular beam experiments this interaction time is typically a few hundred microseconds. The recently demonstrated methods to decelerate and manipulate molecular beams using time-varying electric and magnetic fields [5] enhances significantly the obtainable interaction time and hence resolution. Recently, high resolution microwave spectroscopy was performed on Stark decelerated beams of ammonia [6] and OH [7]. In these proof-of-principle experiments an interaction time of up to a millisecond was obtained. Longer interaction times can be reached in a molecular fountain.

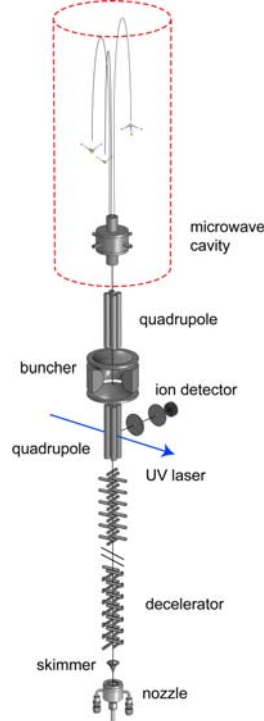


FIGURE 1. Scheme of the molecular fountain.

Figure 1 shows the scheme of the molecular fountain that is currently being tested at the LCVU [8]. In this device a molecular beam of ammonia molecules is decelerated to a few meters per second, cooled and subsequently launched. The molecules fly upwards some 30 cm before falling back under gravity, thereby passing a microwave cavity twice— as they fly up and as they fall back down. The effective interrogation time in such a Ramsey type measurement scheme includes the entire flight time between the two traversals. In this way an interrogation time of 0.5 seconds can be obtained. We use ammonia, NH_3 , as the test molecule in the fountain for a number of reasons: The microwave spectrum of the various isotopomers of ammonia is very well studied. In fact, it was used in the first demonstration of an ‘atomic’ clock by Condon and Lyons in 1948 [9] and the first demonstration of a MASER by Gordon and co-workers in 1955 [10]. Furthermore, ammonia has a strong Stark shift and is easily decelerated using a Stark decelerator. Last but not least, the inversion frequency of ammonia is very sensitive to possible variations of the proton-electron mass ratio.

In the electronic ground state, ammonia has a pyramidal structure with three hydrogen atoms on a plane and the nitrogen atom at the apex. The hydrogen atoms tunnel quantum mechanically through the barrier between the two equivalent configurations of the molecule. As a result, the lowest vibrational energy levels of the molecule are split into a symmetric and anti-symmetric level. The energy difference between the two inversion levels is given simply by Planck’s constant times the tunneling frequency. The tunneling frequency is very sensitive to the mass of the hydrogen atoms as illustrated by the fact that the inversion frequency decreases from 23.8 to 1.59 and 0.3 GHz when the hydrogen atoms in ammonia are replaced by deuterium or tritium atoms, respectively. From these experimental results, it can be inferred that the K-factor of the inversion frequency of NH_3 is equal to -4.6, about ten times more sensitive than ordinary vibrational transitions [8].

In the fountain, it should be possible to measure the inversion frequency with a fractional precision of 10^{-12} – 10^{-14} (depending on the signal-to-noise ratio). By comparing the ammonia clock with the Cesium clock for a period of two years this will result in a constraint for the time-variation of the proton-electron mass ratio of 10^{-13} – 10^{-15} /yr.

2-PHOTON MICROWAVE SPECTROSCOPY OF CO

The enhanced sensitivity in ammonia triggered us to look for possible other mechanisms that lead to an increased sensitivity to the variation of the proton-electron mass ratio in molecular systems. We found that the sensitivity to a variation of the proton-electron mass ratio and the fine-structure constant is enhanced due to near degeneracies between rotational levels in the fine structure ladders of molecular radicals [11]. This is an extension to the work of Flambaum [12] and Flambaum and Kozlov [13]. The enhancement occurs in molecular states that are well described by Hund’s case (a) coupling scheme.

In Hund’s case (a), molecular levels are labeled by the total angular momentum, J , by the projection of \mathbf{L} on the internuclear axis, Λ , by the projection of \mathbf{S} on the internuclear axis, Σ , and by the projections of \mathbf{J} on the internuclear axis, Ω , and on the space fixed axis, M . There are $2S+1$ fine structure levels, characterized by their Ω values. These levels have energies of $A_e\Lambda\Sigma$, with A_e being the spin-orbit constant. Each fine-structure state has a pattern of rotational levels with energies given by $B_eJ(J+1) - D_eJ^2(J+1)^2 \dots$, with B_e and D_e being the rotational constant and the centrifugal distortion constant, respectively, and $J=\Omega$ being the lowest level. In Hund’s case (a), the electronic motion can in first order be separated from the nuclear motion. As a result, the energy differences between the different spin-orbit ladders scale as α^2 but do not depend on μ , whereas the energy differences between different rotational states within a particular spin-orbit ladder depend linearly on μ but do not depend on α . An interesting situation now occurs when two levels of a different spin-orbit ladder have an accidental degeneracy and it is possible to make a (2-photon) microwave transition between these levels. In this case the sensitivity of this microwave transition to a change of μ and α is enhanced by a factor A_e/ν , with A_e being the spin-orbit constant (expressed in Hz) and ν being the frequency of the microwave transition.

Such an accidental degeneracy occurs, for instance, in the metastable $a^3\Pi$ state of CO. In Figure 2, the transitions from $J=4, \Omega=2 \rightarrow J=6, \Omega=1$ and $J=6, \Omega=1 \rightarrow J=8, \Omega=0$ are shown for the most common isotopomers of CO. Here the effects of nuclear spin and hyperfine structure have been neglected. The molecular constants are obtained via isotope scaling of the constants of $^{12}\text{C}^{16}\text{O}$ [14]. The large isotope shift of the transitions is indicative of a large sensitivity to a variation of μ . The solid lines plotted in the figure, follow the relation $E = \text{constant} + \mu_N \times 72.5$ [GHz/amu], with μ_N being the reduced nuclear mass. From this we can determine the sensitivity, $K_\mu = 2\mu_N \times 72.5$ [GHz/amu] / ΔE , with ΔE being the energy difference between the two near degenerate levels. Similar calculations have been performed for the $v=1$ state (not shown).

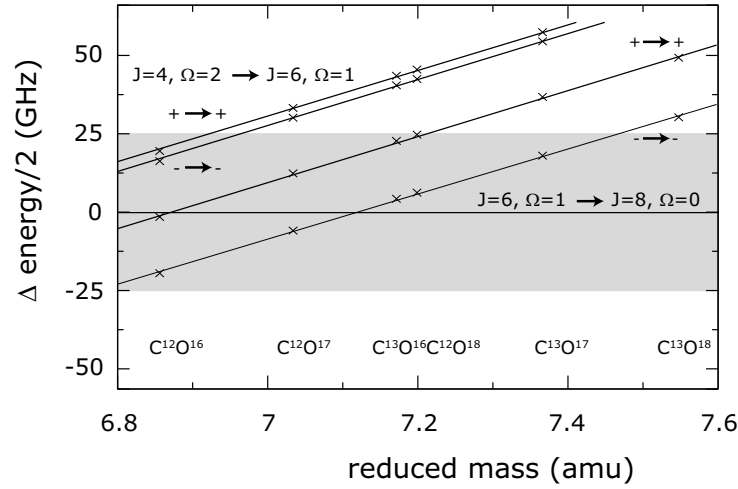


FIGURE 2. Frequencies of selected 2-photon transitions in the $a^3\Pi$ state of (different isotopomers of) CO. The sensitivity for a change of μ is apparent from the large isotope shift of the transition.

We are particularly interested in two transitions in the $v=0$ and $v=1$ of $^{12}\text{C}^{16}\text{O}$ at 1.648 GHz and 2.459 GHz which have a sensitivity to a variation of the proton-electron mass ratio of -302 and 202, respectively. The difference in sign implies that when μ changes, the frequencies of these two transitions will change in opposite directions, i.e., when μ becomes larger, the transition frequency in the $v=0$ will decrease while the transition frequency in the $v=1$ will increase. Combined these two transitions have a sensitivity which is 500 times larger than an ordinary rotational transition. We are currently constructing a molecular beam machine to measure these 2-photon transitions. Ultimately, the precision will be limited by the ~ 5 ms lifetime of metastable CO, but it seems possible to measure the transitions with a fractional precision on the order of 10^{-12} , which would result in a limit for μ -variation on the order of 10^{-14} - 10^{-15} /yr.

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