

Search For New Physics At The Electron-Volt Energy Scale By Precision Spectroscopy Of Cold Molecules

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We present the progress of two experiments that, based on infrared precision laser spectroscopy of either a buffer-gas-cooled or a Stark-decelerated molecular beam, will produce new sets of absolute, ultra-accurate frequency measurements of ro-vibrational lines relative to atomic clock transitions. The ultimate goal is to constrain, over a few-year timescale, the fractional temporal variation of the proton-to-electron mass ratio at a level of $10^{-15}/\text{yr}$. When applied to calculable molecular systems, our approach will furthermore provide a refined grid of line-center frequencies that, combined with sophisticated quantum electrodynamics (QED) computations, might unveil new physical phenomena beyond the Standard Model, including additional long-range hadronic interactions.

Keywords: Proton-to-electron mass ratio; Cold molecular beams, Precision ro-vibrational spectroscopy.

1. Introduction

The Standard Model (SM) of Physics successfully describes every fundamental particle and force at the microscopic scale, yet fails to explain properties of the Universe such as the existence of dark matter, the amount of dark energy, and the preponderance of matter over antimatter. In an attempt to clarify these phenomena, while reconciling perturbative gravity with quantum mechanics within a unified model, String Theory postulates the existence of additional compactified dimensions (whose size is on the order of the Planck

length) where a “fundamental” Lagrangian, containing the “true” constants of Nature, can be defined for elementary objects (leptons, quarks and gauge bosons) [1]. The vacuum expectation values (VEVs) of some a priori massless scalar fields (dilaton and moduli) then settle the factual parameters (effective coupling constants and particle properties) which appear in the “low-energy” limit of the “fundamental” Lagrangian describing our 4-dimensional world, namely the actual Lagrangians corresponding to the 4 known fundamental interactions. In this picture, it is the extent of the extra dimensions to ultimately determine these factual parameters and hence the strength of forces in everyday world [2]. Then, spacetime changes of the effective coupling constants and particle properties are predicted as a direct result of the cosmological evolution of the extradimensional subspace. In a popular theory, this evolution is determined by a dynamical scalar field, dubbed quintessence, that is slowly rolling down a very shallow self-interaction potential. In conclusion, testing the stability of Nature’s fundamental couplings provides a direct handle on new Physics: a detection of variations will be revolutionary, but even improved null results yield competitive constraints on a range of cosmological and particle physics paradigms. 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 targets are the fine structure constant α (defining the scale of quantum electrodynamics) and the proton-to-electron mass ratio $\beta = m_p/m_e$ characterizing the strength of the strong interaction in terms of the electro-weak one. Both parameters determine the energy scaling of atomic/molecular spectra to a certain extent; therefore, if they vary over spacetime, transition frequencies of atoms/molecules taken at two different spacetime points will appear mutually shifted [3]. In particular, the proton-to-electron mass ratio is more accurately addressed with ro-vibrational transitions of molecular systems, where the center frequency scales with β explicitly [4].

2. Experiment in progress

Following the scheme originally demonstrated by Shelkownikov and co-workers [5], our proposal is to measure, over a few-year timescale, the frequency of a suitable molecular ro-vibrational transition relative to a given atomic clock transition. Then, the following relationship between our observable $\omega = \nu_{vib}/\nu_{clock}$ and β is derived:

$$\frac{\dot{\omega}}{\omega} = -0.5 \frac{\dot{\beta}}{\beta} - 2.83 \frac{\dot{\alpha}}{\alpha} - \frac{\dot{\mu}}{\mu}$$

where μ denotes the ratio between the magnetic dipole moment of the atomic nucleus and the Bohr magneton. In combination with constraints on α and β , as inferred from increasingly precise atomic clock frequency comparisons, this can be used to set an upper limit to the temporal variation of β . Our goal is to improve by more than one order-of-magnitude the current record, $\dot{\beta}/\beta = (-3.8 \pm 5.6) \cdot 10^{-14}$, obtained with a conventional supersonic beam of SF₆. To greatly enhance the spectroscopic interrogation time and hence suppress the transit-time broadening, typically setting the ultimate attainable resolution in the line-center frequency measurement, a strategic factor in the experiment will be the realization of a sample of cold stable molecules [6]. In this respect, two different, complementary approaches are being pursued: buffer gas cooling (BGC) and Stark deceleration (SD). Below, preliminary spectroscopic results are presented for each of these two schemes.

2.1. *Cavity ring-down spectroscopy of a buffer-gas-cooled acetylene beam*

By virtue of its applicability to nearly all species and its efficiency in producing very dense samples, the BGC method offers unique perspectives for low temperature precision molecular spectroscopy [7]. However, so far, only electronic molecular transitions have been addressed on BGC beams, either by laser-induced fluorescence (LIF) or resonance-enhanced multi-photon ionization (REMPI), whereas spectroscopy of the weaker, but much narrower, ro-vibrational transitions is still restricted to samples within the BGC cell. In this respect, we recently filled this gap by demonstrating the applicability of a high-resolution and high-sensitivity detection technique, like cavity ring-down spectroscopy (CRDS), on a BGC partially hydrodynamic acetylene beam [8]. Specifically, the performance of our scheme was evaluated for a 10-K C_2H_2 beam on the $g \rightarrow (v_1 + v_3)$ R(1) component at 1.5 μm wavelength using a narrow-linewidth diode laser stabilized against a GPS-disciplined rubidium clock via an optical frequency comb synthesizer (OFCS). As a result, an uncertainty of 330 kHz ($1 \cdot 10^{-9}$ in fractional terms) was achieved in the absolute determination of the line center.

This represents the first step towards the extension of more sophisticated cavity-enhanced interrogation schemes, including saturated absorption cavity ring-down or two-photon excitation, to buffer-gas-cooled molecular beams. In this direction, preliminary sub-Doppler measurements have just been accomplished with room-temperature acetylene, yielding an uncertainty in the line-center frequency determination as low as 2 kHz ($1 \cdot 10^{-11}$ in fractional terms).

2.2. *REMPI-assisted spectroscopy of a slow carbon monoxide beam*

Here, the spectroscopic frequency measurement is performed in a triple-resonance configuration on a slow (318 m/s) metastable CO beam having a density of $10^8/cm^3$ particles in a selected quantum state, similar to what is currently obtained in state-of-the-art setups for cold molecules [9]. First of all, the metastable $a^3\Pi_1, v = 0, J = 1, +$ state is prepared from the ground state with a pulsed laser at 206 nm. Then, a 6- μm -wavelength quantum cascade laser (QCL) drives the ro-vibrational transition to the $v = 1, J = 1, -$ state. From there, the molecules are eventually detected by multiphoton resonance-enhanced ionization. Frequency referencing of the QCL against the Cs primary standard is established via an ultra-stable laser at 1542 nm sent over the Italian fiber-link network [10], the gap between near and mid IR being bridged through a difference-frequency-generation nonlinear process taking place in an orientation patterned (OP) GaP crystal. The stability and robustness of our triple-resonance setup allows for rapid scans providing high signal-to-noise ratios. Indeed, a 25-minutes scan yields a Doppler-broadened absorption profile with a full width at half maximum (FWHM) of 900 kHz, which is already two orders of magnitude narrower than the best value reported in the literature. By acquiring 22 scans over a few days, we could determine the line-center frequency of the chosen ro-vibrational transition as 51399115447 kHz with a fractional uncertainty as low as $6 \cdot 10^{-11}$, improving the current record by more than 4 orders of magnitude.

Also in this case, the resolution of the absolute frequency measurement can be greatly improved after the implementation of an effective SD machine for the CO beam in conjunction with the adoption of a sub-Doppler spectroscopic interrogation technique.

3. Conclusion and perspectives

In the future, application of an ultra-high-resolution spectroscopic technique, like two-photon excitation in the optical domain, to a suitable sample of cold stable molecules, produced either by BGC or SD, will produce new sets of extremely precise frequency measurements of ro-vibrational transitions at the electron volt energy scale. As a prime repercussion, this will serve to put more and more severe constraints on the time variation of the proton-to-electron mass ratio. Furthermore, in the case of calculable molecular systems (especially H₂ and its isotopomers), comparison of line-center frequencies measured by our approach with sophisticated quantum electrodynamics (QED) computations will be used to constrain the coupling strength of additional long-range (internuclear distance) hadron-hadron interactions [11].

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