

Probing m_e/m_p Variation by Comparison of Near-resonant Acetylene Reference Lines at 1.5 μm

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Molecular energy levels are sensitive to electron-to-proton mass ratio constant $\mu=m_e/m_p$ variation which is higher than that of fine structure constant¹. Frequency comparison of two near-resonant rovibrational transitions $f_{1,2}$ in the electronic ground state is discussed to probe variation of μ . The change of the frequency splitting of the transitions $X=f_1-f_2$ with the fractional change of μ : $\partial_\mu X=dX/d(\ln\mu)$ governs the sensitivity of the approach. In addition, measurement of difference frequency reduces some systematic frequency shifts and leads to an uncertainty δX that put the limit of the fractional uncertainty of μ variation at $\delta X/\partial_\mu X$. Finally, when both transitions are measured against a clock at frequency f_c with uncertainty δf_c , the accuracy is no better than $\delta X_{\min}=X\delta f_c/f_c$. Resonant vibrational levels in a polyatomic molecule or isotope effect are exploited to define the pairs of transitions. Near-resonant transitions arise from the cancellation of the difference between the vibrational intervals with a fraction of the difference between the rotational intervals. The gap between the vibrational bands origins yields an enhancement of the sensitivity $\partial_\mu X/X$. The approach of frequency comparison of near-resonant molecular transitions is general and can be extended to rotational transitions in the THz domain.

Isotopic acetylene transitions provide a dense grid of frequency references in the 1.5 μm region. Many groups used a laser locked by the Pound-Drever-Hall technique to saturated absorption lines probed in a Fabry-Perot cavity for absolute frequency measurements² with a frequency comb. Sensitivity of each transition frequency to the fractional variation of μ $\partial_\mu f$ is calculated using a rovibrational Hamiltonian³. Pairs of near-resonant transitions are identified for this data³ and the calculated enhancements are $\partial_\mu X/X>10^4$. The stabilized laser has a fractional frequency stability of a few 10^{-12} at 1 s of integration time with a plateau at some 10^{-14} at 10^3 s. The frequency comparison may put a limit to the μ variation beyond the level reached previously⁴ with careful control of systematic frequency shifts. Pressure, power and modulation lead to fractional frequency shifts in the 10^{-13} range³. The systematic shifts of the difference frequency are reduced and the stability of the experimental parameters defines the ultimate uncertainty. Systematic frequency shifts that have been previously less explored, due to second order Doppler and recoil effects, hyperfine structure and Zeeman effect, black-body radiation shift, are evaluated.

¹ see for example : Eur. Phys. J. Special Topics, vol. 163, 2008; X. Calmet, H. Fritzsch, "The cosmological evolution of the nucleon mass and the electroweak coupling constants", Eur. Phys. J. C, vol. 24, p. 639-642, 2002.

² C.S. Edwards *et al*, "High-accuracy frequency atlas of $^{13}\text{C}_2\text{H}_2$ in the 1.5 μm region", Appl. Phys. B, vol. 80, p. 977-983, 2005 ; C.S. Edwards *et al*, "High-precision frequency measurements of the $\nu_1+\nu_3$ combination band of $^{12}\text{C}_2\text{H}_2$ in the 1.5 μm region", J. Molec. Spectroscopy, vol. 234, p. 143-148, 2005; J. Jiang *et al*, "Measurement of acetylene-d absorption lines with a self-referenced fiber laser frequency comb", J. Opt. Soc. Am. B, vol. 24, p. 2727-2735, 2007.

³ M. Herman *et al*, "Vibrational spectroscopic database on acetylene, $X^1\Sigma_g^+$ ($^{12}\text{C}_2\text{H}_2$, $^{12}\text{C}_2\text{D}_2$, and $^{13}\text{C}_2\text{H}_2$)", J. Phys. Chem. Ref. Data, vol. 32, p. 921-1361, 2003.

⁴ V.V. Flambaum, M.G. Kozlov, "Limit on the cosmological variation of m_p/m_e from the inversion spectrum of ammonia", Phys. Rev. Lett., vol. 98, p. 240801-1-240801-4, 2007.