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Enhanced sensitivity to bosonic ultralight dark matter from acetylene transitions between near-degenerate vibrational modes

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Abstract: Fourier-transform microwave spectroscopy of a transition of $^{12}\text{C}_2\text{H}_2$ with enhanced sensitivity to the proton-electron mass ratio (μ) variation may constrain a fractional variation of μ at the 10^{-12} level in 1 s.

Main body of the paper

The nature of dark matter is unknown despite the work performed in high energy physics towards its direct detection [1]. Couplings between ultralight dark matter candidates to the particles of the Standard Model can induce temporal variations of the fundamental constants that were probed through precision measurements with atoms and molecules [2]. Transitions between near-resonant energy levels of atoms or molecules were addressed in dark matter searches [3] to benefit from their enhanced sensitivity to the variations of the fundamental constants. Acetylene transitions were exploited as frequency references in the 1.5 μm domain. This work addresses the potential for dark matter searches from transitions between near-resonant energy levels of $^{12}\text{C}_2\text{H}_2$.

For this symmetric linear chain molecule, transitions in the microwave and THz domains are enabled by near-resonances of fundamental, harmonics or combinations of the degenerate vibrational bending modes ν_4^{14} at 18.3 THz and ν_5^{15} at 21.9 THz (Fig. 1(a)). The relevant energy levels are modelled using a state-of-the-art Hamiltonian for rotation, centrifugal distortion, vibration, anharmonicity and l-type rotational and vibrational interactions [4]. The parameters of the Hamiltonian are expressed in function of nuclear masses, internuclear distances, vibrational frequencies, and force constants in order to derive the sensitivity of the acetylene energy levels to the proton-electron mass ratio (μ) variation. The relevant microwave transitions of C_2H_2 display enhanced sensitivity coefficients for a variation of μ up to $\pm 10^2$, as it is shown in Fig. 1(b).

Fourier-transform microwave spectroscopy enabled probing molecular spectra in cold supersonic beams at Doppler-limited resolution with high sensitivity [5]. Enabling these performances on transitions between excited vibrational levels may be realised, for example, by probing heated molecular gas in a Fabry-Perot cavity and by optically pumping the population from the relevant rotational level in the vibrational ground state to the lower level of the microwave transition. For example, the transition $(\nu_4^{14}, \nu_5^{15}, J) = (2^{-1}, 0^0, 37) \rightarrow (1^0, 1^0, 36)$ at 10.364 GHz may be interrogated after optically pumping the transition $(\nu_4^{14}, \nu_5^{15}, J) = (0^0, 0^0, 36) \rightarrow (2^{-1}, 0^0, 37)$ in a feasible experimental setup based on C_2H_2 heated at 625 K at 0.24 Pa in a Fabry-Perot cavity (waist radius 10 cm, length 1 m) with 10^5 quality factor. For these parameters, the spectral linewidth of the free induction decay signal is limited

by collisional broadening nearly to the cavity linewidth. The cavity-imposed detection bandwidth is by 10^5 Hz and the signal to noise ratio for $\pi/2$ excitation is 94 per pulse, assuming a noise temperature at 625 K of the heterodyne detection circuit of the molecular signal. Exploiting this signal for frequency measurements may enable access to a stability by 2 Hz at 1 s measurement time. Enhanced sensitivity of this transition allows to constrain a fractional variation of μ at 2×10^{-12} for 1 s integration time.

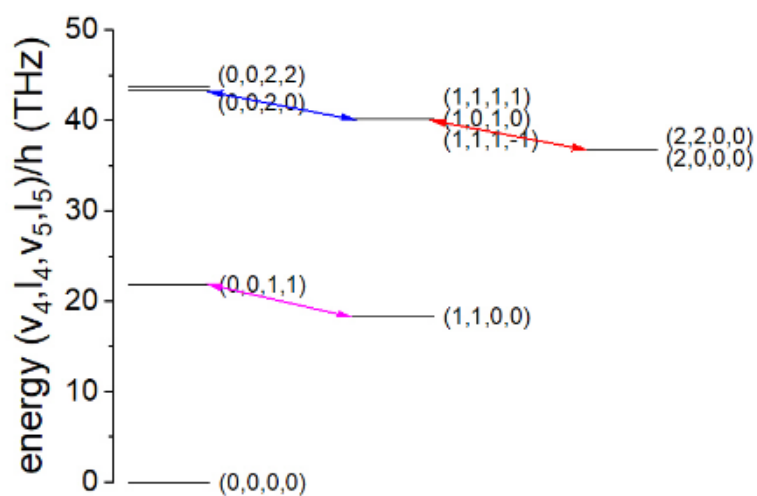
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Figure captions

Fig. 1 (a) The lowest near-degenerate vibrational energy levels of $^{12}\text{C}_2\text{H}_2$. (b) Sensitivity coefficients at a variation of the proton-electron mass ratio for the relevant microwave and THz transitions of $^{12}\text{C}_2\text{H}_2$

(a)



(b)

