Pure frequency-based dispersive spectroscopy

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We explore applicability of several variations of pure frequency-based spectroscopic techniques to molecular systems and their metrology. In these techniques we take advantage from linear phenomenon well-known as mode pushing in an optical cavity with an absorbing medium and intrinsic physical connection between absorption and dispersion. It was demonstrated that mode frequency shifts measurements allow to obtain molecular spectra with exceptional accuracy and precision.

Use of laser tightly PDH-locked to high-finesse cavity filled with absorbing gas allowed scanning of narrow cavity mode resonance and its frequency determination with sub-Hz uncertainties. This technique called cavity mode-dispersion spectroscopy (CMDS), with its primary observable being frequency [1], is immune to bias caused by nonlinearity in the detection of light intensity affecting most commonly used spectroscopic approaches. The CMDS allows molecular transition intensities determination with sub-promille relative uncertainty [2,3] and its traceability to primary frequency standards of the SI. Moreover, CMDS was applied to Doppler-free saturation measurements of weak molecular transitions and appeared to be superior to other techniques [4]. Combination of optical frequency comb as a light source and a Fourier transform spectrometer led to realization of a broadband CMDS [5]. Finally a fast dual-comb detection scheme was implemented to CMDS [6,7]. Even faster realization of these approaches is possible by beating light buildup or decaying from the optical cavity with a local oscillator precisely detuned from the cavity resonance. We demonstrated the cavity buildup dispersion spectroscopy (CBDS) [8] allowing rapid measurement of the mode frequency on a time scale shorter than the cavity decay time and a broadband experiment using dual-comb cavity ring-down spectroscopy (DC-CRDS) [9].

The pure frequency-based dispersive spectroscopy seems to be an attractive alternative to intensity-based measurements of cavity decay rates or light absorption, especially in studies of weak molecular transitions. Applications include reference data for a new generation of spectroscopic databases, studies of fundamental physics, gas metrology and Doppler width thermometry.

References

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