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Saturation Spectroscopy of Rotational Transitions Toward the Molecular Clock Application

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Rotational transitions of polar molecules give rise to molecule-specific spectra in the terahertz (THz) region and have long been used in chemical analysis and the search for interstellar molecules. Most of these studies rely on linear absorption spectroscopy, where the absorption linewidth is dominated by Doppler broadening resulting from the thermal motion of the molecules. Since the rotational levels of molecules are not equally spaced, specific transitions can be isolated and treated as two-level systems. In addition, their transition frequencies are remarkably stable against external stimuli, such as electromagnetic fields, rendering them suitable candidates for frequency standards known as “molecular clocks.” Recently, the IMRA group demonstrated that employing a dual-wavelength Brillouin laser as the THz source—stabilized to the OCS rotational transition at 316 GHz—yielded a fractional frequency instability of $1.2 \times 10^{-12}/\sqrt{\tau}$ [1]. However, such approaches are fundamentally limited by the inevitable Doppler linewidth. One effective method to overcome this limit is saturation spectroscopy, such as Lamb-dip spectroscopy.

In this study, we quantitatively characterize the absorption saturation effect and determine the saturation parameters for rotational transitions in acetonitrile molecules. Furthermore, we discuss the potential application of Lamb-dip spectroscopy to frequency stabilization and the development of high-precision molecular clocks. Lamb-dip spectroscopy was performed using a frequency-stabilized sub-THz light source [2]. A typical Lamb-dip spectrum is shown in Fig. 1. From the fitting analysis, the transition frequency was determined with an accuracy of 10^{-9}.

Furthermore, we constructed a differential spectroscopy system and attempted to stabilize the frequencies of the free-running diode lasers used for difference-frequency generation in the sub-THz light source by using molecular rotational transitions. In the absence of the Lamb-dip (i.e., using the Doppler-broadened line), we achieved a frequency stability of 1.0×10^{-9} at an integration time of $\tau = 1$ s. We will also discuss the potential of using the Lamb-dip to further enhance frequency stability.

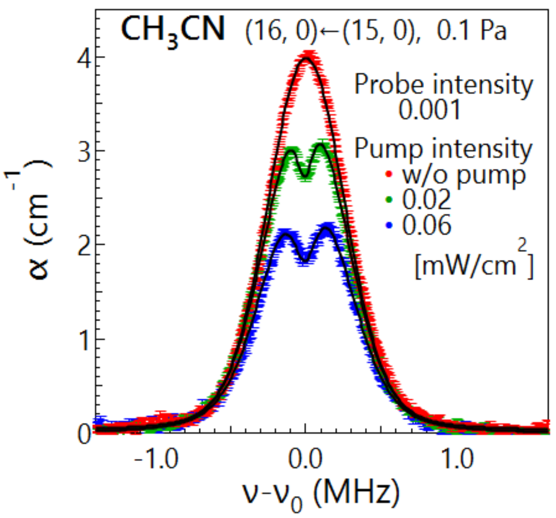

Fig. 1. Pump intensity dependence of Lamb-dip spectra of acetonitrile molecules at 0.1 Pa. ν_0 =294302.389 MHz.

REFERENCES

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