LOW-FREQUENCY DETECTION MICROWAVE THREE-WAVE MIXING

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Microwave three-wave mixing, which was first demonstrated in 2013 by Patterson, Schnell, and Doyle [1, 2] and described eloquently in NMR terminology by Grabow [3], has shown its applicability in differentiating enantiomers as well as quantifying enantiomeric excess for chiral molecules in the gas phase. Further theoretical development of rotational three-wave mixing has been presented by Lehmann. [4] However, this technique's capability has not been fully explored and the sensitivity of different measurement implementations have not been evaluated. This work presents a simplified measurement scheme in which two high-frequency (4-12 GHz) transitions of a chiral gas sample are excited with orthogonally polarized pulses of light utilizing a dual-polarization wave horn antenna. The low-frequency chiral emission signal (600-2000 MHz) is detected at the mutually orthogonal polarization and perpendicular to the excitation pulse propagation direction. In this measurement scheme, the detection electronics and digitization rate required are simplified and reduced. Propylene oxide, alanine, and menthone introduced in a pulsed-jet expansion are used to demonstrate the low-frequency detection method and its comparison to traditional three-wave mixing schemes where detection of a high-frequency signal is employed. Additionally, some other nuances of the measurement technique have been explored including the angular dependence of the chiral emission and the importance of the polarization in the excitation and detection horn antennae.

[1] D. Patterson, M. Schnell, and J.M. Doyle, "Enantiomer-specific detection of chiral molecules via microwave spectroscopy", Nature 497, 475-478 (2013). [2] D. Patterson and J.M. Doyle, "Sensitive Chiral Analysis via Microwave Three-Wave Mixing", Phys. Rev. Lett. 111, 023008 (2013). [3] J. Grabow, "Fourier Transform Microwave Spectroscopy: Handedness Caught by Rotational Coherence", Angew. Chem. Int. Ed. 52, 11698-11700 (2013). [4] K.K. Lehmann, "Theory of Enantiomer-Specific Microwave Spectroscopy", in Frontiers and Advances in Molecular Spectroscopy, 713-743 (2018).