IR/THZ DOUBLE RESONANCE SIGNATURES AT ATMOSPHERIC PRESSURE

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IR/THz double resonance (DR) spectroscopy, historically used to investigate molecular collision dynamics and THz molecular lasers at low pressures ($<$ 1 Torr), shows promise for trace gas remote sensing at atmospheric pressure. Molecular specificity is obtained through the rare coincidence(s) between molecule-specific ro-vibrational energy levels and CO_2 laser lines. The resulting molecule-specific, DR-induced, THz spectroscopic signatures strongly depend on the type of ro-vibrational transition involved (P, Q, or R), the type of vibrational level excited (stretching or bending), and the molecular mass. To illustrate these sensitivities, calculated DR spectra of prototypical molecules such as methyl fluoride, methyl chloride, and methyl cyanide will be discussed. Although atmospheric pressure broadening obfuscates pure rotational spectra, we show how it can enhance the DR signature in two ways: by relaxing the pump coincidence requirement and by adding the DR signatures of multiple nearby transitions. We will present estimates of this enhancement, including cases where the coincidences that produce the strongest DR signatures at atmospheric pressure do not exist at low pressures.