DETECTION AND INTERPRETATION OF COLLISIONAL TRANSFER AND ROTATIONAL ANISOTROPY FINGERPRINTS IN RESONANT FOUR-WAVE MIXING SPECTRA.

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Coherent responses produced by resonant four-wave mixing (RFWM) in a weakly absorbing medium carry valuable information on the intrinsic properties and dynamics of the quantum states involved. Here, two aspects of RFWM applications are highlighted. First, the Two-Color (TC) version of RFWM was found to be a unique spectroscopic tool to directly trace collisional state-to-state transfer in isotropic gaseous media, both in the frequency\(^a\) and time\(^b\) domains. Second, the RFWM techniques appeared to be very useful for studies of the rotational anisotropy\(^c\). Here we report new experimental one-color RFWM spectra of the OH radicals produced by laser photolysis of H\(_2\)O\(_2\) at 266 nm. Polarization dependence and Doppler line structure of the spectra show clear evidence of the pronounced anisotropy of angular momentum (\(j\)) and velocity (\(v\)) distributions as well as on the \(j\)-\(v\) correlation. The obtained results directly point to the pronounced OH helicity (i.e. \(j\parallel v\)) which yet remained beyond the reach of purely optical means. For all mentioned cases, the line-shape theory\(^d\) is an optimal tool to derive compact expressions for the RFWM signals.

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