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Polarization- and time-resolved DFWM spectroscopy for studies of transient anisotropy of molecular motion

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Photo-dissociation of H_2O_2 molecules by linearly polarized ns-pulse radiation at 266 nm results in transient anisotropies of nascent OH fragments recoil velocity (\mathbf{v}) and angular momentum (\mathbf{J}) vector distributions. These anisotropies have been probed at arbitrary time delays after the H_2O_2 photo-dissociation pulse using degenerate four-wave mixing (DFWM) with appropriately adjusted polarizations of pulsed input and signal beams. DFWM lineshapes of rovibronic transitions in the $A^2\Sigma^+ \leftarrow X^2\Pi$ (0,0) band of nascent OH have been recorded.

The polarization-resolved DFWM spectra show clear evidence of vector correlations between the parent H_2O_2 molecule transition dipole moment, the OH fragments recoil velocity and angular momentum. The variations of DFWM line shapes and intensities with the time delay characterize collisional disturbance of $\mathbf{J}\text{-}\mathbf{v}$ correlation and relaxation of rotational and translational anisotropies.

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