INVESTIGATION OF TEMPERATURE-DEPENDENT COLLISION DYNAMICS OF GASEOUS MOLECULES WITH TIME-RESOLVED ULTRAFAST CARS

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Temperature-dependent collision dynamics of nitrogen and hydrogen molecules are investigated using time-resolved ultrafast coherent anti-Stokes Raman scattering (CARS). This study has profound potential impact on the development of density-matrix-based models for extracting temperatures from measured ultrafast CARS signals. There are four promising advantages of short-pulse CARS spectroscopy: (1) it improves accuracy by reducing or eliminating the non-resonant contribution to the CARS signal when the probe beam is delayed with respect to the pump beam, (2) it minimizes the effect of collisions on the CARS signal, thereby reducing modeling uncertainty and increasing signal-to-noise ratio, (3) it improves sensitivity and might enable the detection of minor species through reduction or elimination of interference from the non-resonant background, and (4) it has the capability of generating signal at rates up to 1 kHz. These advantages have the potential to enhance the performance of CARS thermometry in high-pressure, high-temperature combustors of practical interest by overcoming known limitations of nanosecond-based systems.