POLARIZATION FEATURES OF THIRD-HARMONIC GENERATION FROM COHERENTLY SPINNING MOLECULES

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The polarization of ultraviolet short pulses produced by third-harmonic generation (THG) in a gas of coherently spinning molecules is investigated. A pulse of twisted linear polarization is used to produce a unidirectional rotational motion of molecules\(^1\). The THG is driven by a second pulse, time delayed and circularly polarized in the plane of rotation of the molecules. This fundamental radiation produces two Doppler-shifted harmonics fields of opposite circular polarization\(^2\).

A time-dependent analysis of the THG polarization based on the measurement of the Stokes parameters reveals that the coherently spinning molecules allow the generation of a well polarized third-harmonic field with a high degree of ellipticity. The rotation of the molecular axis can be deduced from the orientation of the polarization ellipse determined by scanning the delay between the two pulses. This enables the determination of the average angular velocity of the molecules. The present method provides a user-friendly polarization-based tachometer for optical detection of spinning nonlinear rotors\(^4\).


