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Title: Generation of ultrashort laser pulses by high-order stimulated Raman scattering and four-wave mixing

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Transient stimulated Raman scattering (TSRS) in hydrogen gas is utilized for generating ultrashort near-infrared pulses, where two gas-cells are utilized to lead to a high efficiency in TSRS. A 100-fs near-infrared pulse is compressed to a sub-20 fs duration. Pulse compression based on molecular phase modulation is also investigated. A pump pulse longer than a molecular rotational period is used to induce a coherent molecular rotation through TSRS. The frequency of a time-delayed probe pulse is modulated by the coherent molecular rotation. A train of 23-fs pulses is obtained with an energy exceeding 10-mJ in this study. For generating an isolated ultrashort pulse, two types of coherent molecular rotations are utilized in the molecular phase modulation. After optimization of the time separation between the pump and probe pulses, the intensities of subpulses are decreased, and a 11-fs near-ultraviolet pulse is generated. Self-phase modulation is also combined with molecular phase modulation to decrease further the intensity of subpulses, which is also necessary to generate a short pulse with a higher pulse energy. For inducing a coherent molecular motion efficiently, a polarization-shaped pulse is used. The induction efficiency of coherent molecular motions by the polarization-shaped pulse is higher than that by conventional TSRS. Recently, generation of sub-10-fs deep-ultraviolet pulses is investigated. In this study, broadband chirped-pulse four-wave mixing is used to lead to an excellent temporal profile in the compressed temporal profile. The deep-ultraviolet pulse is suitable for ultrafast spectroscopy in biologically relevant molecules such as nucleobases and DNA.

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