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Extreme rotational excitation with long sequences of intense femtosecond pulses MARTIN BITTER, VALERY MILNER, University of British Columbia — We present an experimental approach to rotational excitation of molecules capable of creating ultra-broad rotational wave packets inaccessible with other methods, including the technique of an optical centrifuge. Our approach is based on an impulsive excitation by a long sequence of more than 20 laser pulses with peak intensities within each pulse exceeding 10^{13} W/cm². The method overcomes the three obstacles on the way towards extreme rotational excitation: (i) the strong-field effects associated with a single-pulse scheme, (ii) the centrifugal distortion and Anderson localization in a multi-pulse approach, and (iii) the bandwidth limitation of an optical centrifuge. In oxygen, we demonstrate the ability to populate rotational states with an angular momentum $N \approx 250\hbar$, more than twice higher than previously achieved with the centrifuge. Precise timing of the pulses and their spectral broadening due to molecular phase modulation, essential to this technique, are demonstrated and discussed.

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