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High Degrees of Impulsive Alignment in Repetitively Excited N_2 at STP[1] JAMES CRYAN, RYAN COFFEE, PHILIP BUCKSBAUM, The PULSE Institute for Ultrafast Energy Science, SLAC National Accelerator Laboratory — We demonstrate a high degree of both transient and time-independent alignment in Nitrogen at STP resulting from multiple impulsive Raman excitations with linearly polarized light. The alignment is optimized by exploiting the structure of the density matrix, $\rho(J, m_J)$. Our experiment demonstrates a time-independent population alignment, defined as the time average of $\langle \cos^2 \theta \rangle$, that exceeds the single pulse transient coherent alignment. We compare our experimental results to a quantum calculation, which suggests that transient alignment following multiple excitations can exceed $\langle \cos^2 \theta \rangle \sim 0.6$. Under impulsive excitation the entropy and quantum purity remain constant, but both the energy of the ensemble and the J-state distribution move markedly away from a thermal distribution. Transient alignment is related to rotational coherence $C_2 = \left(1 - \frac{\mathrm{tr}(\mathrm{diag}(\rho)^2)}{\mathrm{tr}(\rho^2)}\right)^{1/2}$. We show that this C_2 coherence grows monotonically with our train of eight impulses.

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