High Degrees of Impulsive Alignment in Repetitively Excited N$_2$ at STP

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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 $C_2 = \left( 1 - \frac{\text{tr(diag}(\rho^2)}{\text{tr}(\rho^2)} \right)^{1/2}$. We show that this $C_2$ coherence grows monotonically with our train of eight impulses.

1This research is supported by the US DOE Office of Basic Energy Science, through the PULSE Institute.

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Date submitted: 24 Jan 2009

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