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Adaptive strong-field control of vibrational population in NO^{2+}[1]

E. WELLS, O. VOZNYUK, ADAM BROIN, R. AVERIN, Department of Physics, Augustana University, Sioux Falls, SD 57197 USA, BETHANY JOCHIM, M. ZOHRABI, K.D. CARNES, I. BEN-ITZHAK, J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, KS 66506 USA — An adaptive closed-loop system incorporating coincidence time-of-flight feedback is used to determine the optimal intense laser pulse shapes for manipulating the branching ratio of NO^{2+}. Selection between the NO^{2+} and $N^+ + O^+$ final products requires control of the vibrational population distribution in the transient NO^{2+}, with $v \geq 12$ of the NO^{2+} X $^2\Sigma^+$ state dissociating to $N^+ + O^+$. The ability to both suppress and enhance NO^{2+} relative to $N^+ + O^+$ is observed, with the effectiveness of shaped pulses surpassing near Fourier transform-limited pulses by about an order of magnitude in each direction, depending on the pulse energy. The control is subsequently investigated using velocity map imaging, identifying plausible dissociation pathways leading to $N^+ + O^+$. Combining this analysis with a well-defined control objective supports the conclusion that the primary control mechanism involves selectively populating non-dissociative NO^{2+} vibrational states. The optimized pulse complexity increases as the laser intensity increases, complicating determination of the molecular dynamics underlying this control.

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