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LeRoy Apker Award Lecture: Strong-field dissociation dynamics of NO²⁺: A multiphoton electronic or vibrational excitation?¹

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A 3-D momentum imaging technique is employed to study intense ultrafast laser-induced dissociation of a metastable NO²⁺ beam. We focus on N⁺+ O⁺ coincidences and explore possible dissociation pathways using estimates of the initial vibrational population and transition rates between the X ²Σ⁺ and A ²Π states together with our measured kinetic energy release and angular distribution spectra. Our analysis suggests that lower intensity pulses (<10¹⁵ W/cm²) drive perpendicular transitions between these states. Higher intensity pulses (~10¹⁶ W/cm²), on the other hand, yield a prominent contribution from molecules breaking parallel to the polarization. An intriguing possibility is that this feature is due to a two photon permanent dipole transition to the vibrational continuum of the X ²Σ⁺ state, *i.e.*, a multiphoton vibrational excitation involving only the electronic ground state. The results of our time-dependent Schrödinger equation calculations comparing the probabilities of this type of pathway and competing electronic transitions will be presented.

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