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# Femtosecond photoelectron imaging of NO at 410 nm

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ABSTRACT

The simultaneous measurement of the photoelectron kinetic energy and angular distributions of NO were performed as a function of 410 nm laser intensities by femtosecond photoelectron imaging. The photoelectron kinetic energies are nearly independent of the laser intensities. The insensitivity of the photoelectron kinetic energy to the laser peak intensity suggests that peaks in the photoelectron spectra result from Freemen resonance, characteristic of the occurrence of real population on the intermediate states. The relative amplitudes of photoelectron peaks indicate that lower laser intensities emphasize the resonant population while higher laser intensities favor nonresonant population. The $A^2\ \Sigma^+(\upsilon=2)$ state can be weakly coupled to the $B^2\Pi(\upsilon=4)$ state, results in a small part of population on $B^2\Pi(\upsilon=4)$ state transferred from $A^2\ \Sigma^+(\upsilon=2)$ state. The strong field effect leads to more anisotropy of ATI than that of non-ATI. The modulation of populations in electronic states can be achieved by adjusting laser peak intensities. The results can provide some important basis for realizing quantum manipulation of molecules experimentally.


## 1. Introduction

The multiphoton ionization (MPI) dynamics of nitric oxide is in intense femtosecond laser fields has been studies extensively [1–14]. The ac Stark effect was studied quantitatively using fluorescence spectra and theoretical calculations [1–3]. The field modulation of populations in electronic states of NO was studied experimentally by femtosecond time-resolved photoelectron spectroscopy in an intense two-color laser field [4–7]. Ludowise et al. [5] suggested that the Rydberg-valence coupling strength between the $B^2\Pi$ and $C^2\Pi$ states changes with increasing laser intensity, results in the difference of photoelectron energy spectra for different delay times and pump intensities, which was further confirmed theoretically by Meng et al. [8]. Wang et al. [6] presented that populations in the electronic states exhibited time-dependent behaviors using femtosecond photoelectron imaging in a bichomatic laser field. They observed experimentally the weak coupling between the $A^2\ \Sigma^+$ and $B^2\Pi$ states, which was also referred to using fluorescence spectra [3] and theoretical calculations [2,9]. Wang et al. [10] suggested that the Rydberg–Rydberg Raman coupling and quantum interference between different pathways affect the peak height in the photoelectron energy spectra and the ionization probability of NO. Luque and Crosley [11] determined the electronic transition moment and transition probabilities of the NO $A^2\ \Sigma^+ - X^2\Pi$ and $D^2\ \Sigma^+ - X^2\Pi$ bands by dispersed fluorescence scans. Tsubouchi and Suzuki [12] measured the laboratory frame photoelectron angular distributions (LF-PAD) of $A^2\ \Sigma^+$ state using femtosecond time-resolved photoelectron imaging. But the LF-PAD is a quantity highly averaged over a wide intensity range, leading to less structured than the photoelectron angular distribution in the molecular frame (MF-PAD)

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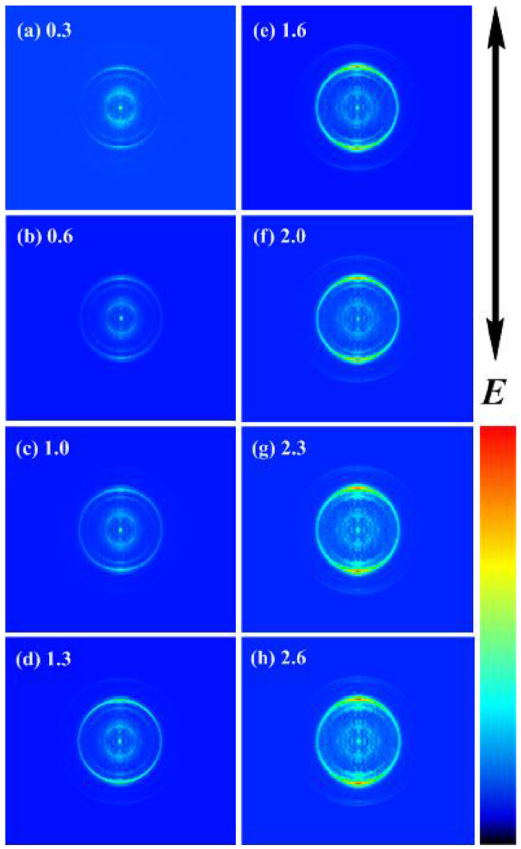


**Fig. 1.** Inverted photoelectron images of NO at eight laser intensities. Intensities are indicated in unit of $10^{14}$ W/cm$^2$. The vertical arrow and the gray bar represent the laser polarization and signal intensity, respectively.

[13]. Although complete explanation of LF-PAD is still unachievable, it can provide helpful information for the ionization channel assignment. Cooper and Zare [14] predicted that the angular distribution of *s*-type electron shows more anisotropy than that of a non-*s*-type electron.

Most experimental and theoretical studies above on MPI of NO are based on photoelectron spectra obtained in a two-color laser field [4–10,12,13]. No work was reported based on the femtosecond single-color imaging. This paper presents new data on the photoelectron dynamics of NO in 70 fs, 410 nm laser fields within MPI region below $3.0 \times 10^{14}$ W/cm$^2$ using velocity mapping technique. The kinetic energy and angular distributions of proton at different intensities are determined. The possible ionization processes are proposed and discussed.

## 2. Experimental set up

The experimental setup is similar to the setup described in Refs. [6,15,16]. Briefly, our homemade solid-state femtosecond laser system produces 820 nm fundamental pulses with 70 fs pulse duration and 20 Hz repetition rate with 160 mW average powers. Pulse duration is monitored with an interferometric autocorrelator. The fundamental output is frequency doubled by a $\beta$-$BaB_2O_4$ (BBO) crystal to yield second harmonic generation (SHG) light, centered around 410 nm.

The SHG beam is focused by a lens of 30 cm focal length into the vacuum chamber of time-of-flight mass spectrometer (TOF-MS). The laser intensities can be varied by neutral density attenuators. The laser peak intensity is calibrated by stark shifts in photoelectron energy spectra of Xe, and are estimated to vary from $0.3 \times 10^{14}$ W/cm$^2$ to $2.6 \times 10^{14}$ W/cm$^2$ in our experiment. The laser polarization direction is perpendicular to the TOF axis.

The sample gas, 5% NO in He, is expanded into the ionization accelerating region through a pulsed valve. After a 40 cm field-free region, electrons are projected onto a two-stage multichannel plate (MCP) detector backed by a phosphor screen. Images on the screen are captured with a computer based charge-coupled device (CCD) camera. Each image is the integration over 20 000 laser shots. The emission from the phosphor screen is monitored by a photo-multiplier tube. Energy and angular distributions are obtained by applying an Abel inversion procedure to the raw images [17,18]. With molecular beam on, the source chamber and flight chamber are maintained at $4.5 \times 10^{-4}$ Pa and $8 \times 10^{-6}$ Pa to avoid the space-charge effect.