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To cite this article: Joshua Beck *et al* 2017 *J. Phys.: Conf. Ser.* **875** 032021

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## Intense-Field Photoionization of Molecules using Ultrashort Radiation Pulses: REMPI in Toluene, Aniline, Phenol, and Fluorobenzene

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**Synopsis** We investigate the intense-field photodynamics of molecules using 50-fs pulses with intensities of  $10^{12}$  to  $10^{14}$  W/cm<sup>2</sup> at 800-nm from a Ti:sapphire laser. We use a mass spectrometer that avoids focal intensity averaging. We have investigated the isoelectronic target molecules toluene (C<sub>6</sub>H<sub>5</sub>-CH<sub>3</sub>), aniline (C<sub>6</sub>H<sub>5</sub>-NH<sub>2</sub>), phenol (C<sub>6</sub>H<sub>5</sub>-OH), and fluorobenzene (C<sub>6</sub>H<sub>5</sub>-F). We discuss a resonance-enhanced multiphoton ionization (REMPI) mechanism that includes the AC Stark shifts of the molecular states. We have also investigated CO<sub>2</sub> and CS<sub>2</sub>. For these molecules, no resonance effects are observed.

We report on the ultrafast (50 fs), intense-field ( $10^{12}$  to  $10^{14}$  W/cm<sup>2</sup>) photoionization and fragmentation of molecules. We investigated a series of four isoelectronic monosubstituted aromatics: toluene (C<sub>6</sub>H<sub>5</sub>-CH<sub>3</sub>, *IP* = 8.83 eV), aniline (C<sub>6</sub>H<sub>5</sub>-NH<sub>2</sub>, *IP* = 7.72 eV), phenol (C<sub>6</sub>H<sub>5</sub>-OH, *IP* = 8.51 eV), and fluorobenzene (C<sub>6</sub>H<sub>5</sub>-F, *IP* = 9.20 eV). Data is recorded under intense-field, single-molecule conditions. We focus 50-fs, 800-nm radiation pulses into a dilute target vapor, and record ion mass spectra for intensities of  $10^{12}$  to  $10^{14}$  W/cm<sup>2</sup>. Our ion mass spectrometer has an interaction volume of micrometer size in all three dimensions, which allows us to record ion yields without focal intensity averaging, i.e., for sharply determined intensities [1]. This also allows us to estimate absolute ionization probabilities.

For the wavelength and intensities we use, multiphoton ionization is expected to dominate over tunneling ionization (Keldysh parameter > 1). If the smallest number of photons needed to reach the ionization continuum is *N*, the ion yield is expected to increase with intensity as  $I^N$ , at least for intensities low enough to avoid channel closing due to ponderomotive shifts. Remarkably, the ion yields of toluene, aniline, phenol, and fluorobenzene do not follow this simple integer-power dependence. For increasing intensities, but before saturation sets in, the ion yield's intensity dependence changes distinctly from  $I^N$  to a lower power of the intensity.

We believe this change is evidence of a resonance-enhanced multiphoton ionization (REMPI) pathway. By fitting our data we find that the lower power is typically an integer, which we interpret as the multi-photon order of the rate-determining step. We had previously seen signatures of molecular REMPI in the monosubstituted halobenzenes C<sub>6</sub>H<sub>5</sub>-X (X = F, Cl, Br, I).[2] Guided by correlations between the excited states of isoelectronic molecules we propose candidates for the molecular resonant states.

To further investigate the role of resonances in molecular intense-field ionization we have started experiments at longer wavelengths, using an optical parametric amplifier source (TOPAS). Work in progress will be discussed.

This material is based upon work supported by the National Science Foundation under Grant No. PHY-0855675 and by the National Science Foundation EPSCoR RII Track-2 CA Award No. IIA-1430519 (Cooperative Nebraska-Kansas grant).

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