

Intense-Field Ionization of Cyclic Hydrocarbon Molecules Measured with Spatial Resolution of Focal Ion Distributions

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Synopsis: We present experimental results of fully three-dimensionally resolved ion yields for benzene and benzene-like molecules, including some heterocyclic compounds. Full spatial resolution of the ion yields allows circumvention of the “volume effect”, in which ions are collected from the entire laser focus at a variety of intensities. As a result of avoiding this effect, our ion yields very closely resemble the true ionization probability, and have allowed us to clearly measure resonance-enhanced multiphoton ionization processes (REMPI).

We present experimental results of spatially-resolved ion densities of benzene and other benzene-like molecules through the use of a “photodynamical test tube” which circumvents volumetric weighting of the laser focus (the volume effect) [1]. Within the volume of the test tube the laser intensity is essentially constant, resulting in ion yield measurements very near to the true ionization probability of the target molecule.

Using laser radiation with a central wavelength of 800 nm and pulse duration of 50 fs focused to intensities of 10^{13} to 10^{15} W/cm², we recorded experimental evidence of resonance-enhanced multiphoton ionization (REMPI) processes in benzene, fluorobenzene, chlorobenzene, bromobenzene, and iodobenzene. For all these molecules we observe a six-photon process and a three-photon resonance, which we believe to be the result of the saturation of an intermediate resonant state ($S_0 \rightarrow S_1$).

In addition, we present results of the ionization and fragmentation of the six-member-ring aromatic hydrocarbons toluene ($C_6H_5-CH_3$), aniline ($C_6H_5-NH_2$), nitrobenzene ($C_6H_5-NO_2$), and phenol (C_6H_5-OH), as well as three five-member-ring aromatic heterocycles: thiophene (C_4H_4S), furan (C_4H_4O), and pyrrole (C_4H_4NH). These molecules display a richer behavior than the halogenated benzenes mentioned above; we believe we have observed effects such as a lowered continuum in aniline, a possible shifted resonant intermediate state in phenol, and multiple saturated resonant states in furan. Recent experimental progress and theoretical interpretations will be presented.

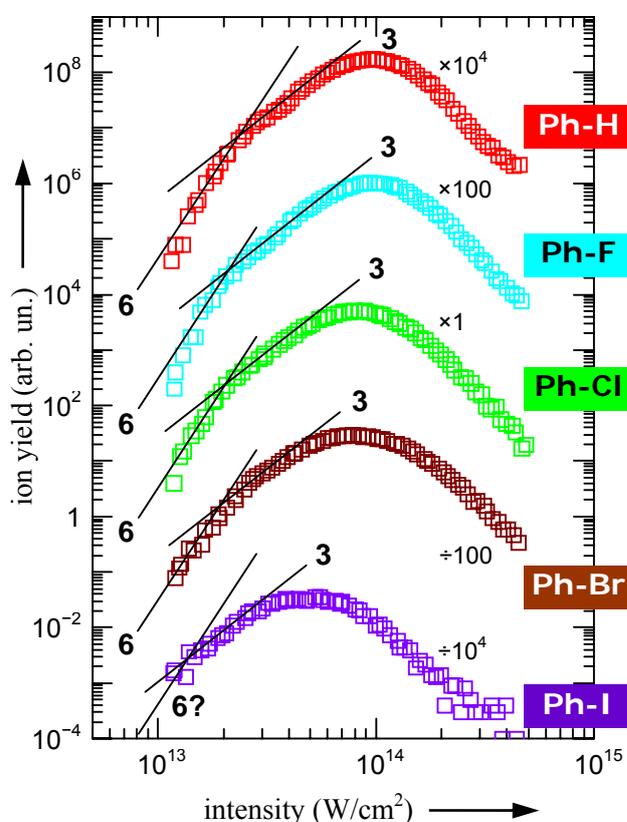


Fig. 1. Measured ion yields of the halogenated benzene series showing (3+3) REMPI processes. The ion yields have been recorded without integrating over the focus.

Reference

[1] J. Strohaber and C.J.G.J. Uiterwaal, “*In situ* measurement of three-dimensional ion densities in focused femtosecond pulses,” *Phys. Rev. Lett.* **100**, 023002 (2008). — Highlighted in *Science* **319**, 699 (Editor’s Choice, 8 Feb. 2008).

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