Abstract Submitted
for the APR05 Meeting of
The American Physical Society

Structure sensitive photoionization via Rydberg levels NARAYANAN KUTHIRUMMAL, College of Charleston, PETER WEBER, Brown University — We introduce Rydberg level spectroscopy as a new method to fingerprint molecular structures, and demonstrate it on the example of the azulene/naphthalene system. To access the Rydberg levels and measure their electronic energies, we employed the double resonant, one- color, 3-photon ionization scheme using 266 nm, 180 fs laser pulses. The observed Rydberg spectra show a rich set of peaks in the range of principal quantum numbers $n = 3$ and 5. The corresponding quantum defects have also been calculated. The results indicated that the present technique has the useful attributes of sensitivity toward large-scale molecular structure features, universal applicability from small molecules to large composite or polymeric systems, and the ease of combination with traditional mass spectrometry. We have performed similar experiments on several other molecules, including phenol, toluene, and various fluorophenols. We found that in all cases the photoionization experiment gives rise to molecule-specific Rydberg spectra. We conclude that the ionization via the Rydberg levels is a generally applicable scheme that can provide fingerprints of the molecular shape during the ionization process.

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Date submitted: 19 Jan 2005 Electronic form version 1.4