DYNAMICS OF DISSOCIATIVE RECOMBINATION, ELECTRON LOSS AND ISOLATED-CORE PHOTON ABSORPTION IN SINGLE ROVIBRONIC RYDBERG RESONANCES OF ^{11}BH

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Optical-optical-optical triple resonance spectroscopy isolates transitions to vibrationless Rydberg states with principal quantum numbers from $n = 7$ to 50 converging to the lowest ionization threshold of ^{11}BH. These transitions appear in the excitation spectrum of photoionized B* atoms produced by dissociative relaxation of ^{11}BH* Rydberg resonances. We observe a region in the spectra that shows evidence for further step of photoabsorption by the BH$^+$ core that leaves the quantum numbers of the Rydberg electron unchanged. For a certain range of third-photon excitation frequencies, a strongly allowed $\mathrm{A}^2\Pi \leftarrow \mathrm{X}^2\Sigma^+$ valence electronic absorption by the ion occurs, for which the initially prepared Rydberg electron acts as a spectator. Using a simple mathematical model in which we allow only transitions vertical in n, we show that the interval of n for which this $|\mathrm{A}^2\Pi\rangle|nl\rangle \leftarrow |\mathrm{X}^2\Sigma^+\rangle|nl\rangle$ isolated-core excitation (ICE) rate exceeds that of the predissociation of ^{11}BH* depends on the finite linewidths and positions of $|\mathrm{A}^2\Pi\rangle|nl\rangle$ features and on the n-dependent predissociative lifetimes of ^{11}BH* Rydberg molecules. Despite relatively broad resonance features, final states are found to decay slowly to neutral products, and, above threshold, where predissociation competes with direct ionization, the rate of electron loss exceeds neutral fragmentation by only an order of magnitude. Interference lineshapes observed for purely predissociative $v^+ = 0$ Rydberg states match closely with states of the same principal quantum number built on $v^+ = 1$, which yield only the ionic product, ^{11}BH$^+$. We conclude that inelastic cation-electron interactions in ^{11}BH proceed via coupling to a common continuum which gives rise to neutral and ionic products.

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