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Macrodimers and Long-Range Rydberg Molecules¹

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The large polarizability of atoms in highly excited states, so called Rydberg states, leads to strong and long-ranging interactions between such atoms. Interacting pairs of Rydberg atoms represent a very exotic molecular system, characterized by high internal excitation, high density of electronic states, internuclear separations exceeding one micrometer, and lifetimes beyond tens of microseconds. I will discuss the computational methods we have developed to determine the electronic structure of interacting Rydberg-atom pairs [1] and our spectroscopic approaches to verify these calculations.

Recently, we could observe the formation of macrodimers, vibrational bound states of two interacting Rydberg atoms, which were predicted in 2002 by Boisseau and coworkers [2,3]. I will discuss the employed sequential photoassociation scheme and the validity of the Born-Oppenheimer approximation for these states.

In an extended outlook, I will discuss our current and planned experiments towards determining electron-atom-scattering phase shifts from high-resolution spectroscopy of another class of exotic molecules, long-range Rydberg molecules where a Rydberg atom and a ground-state atom located inside the Rydberg orbit are bound to each other [4].

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[3] C. Boisseau, I. Simbotin, and R. Côté, Phys. Rev. Lett. 88, 133004 (2002)

[4] C. H. Greene, A. S. Dickinson, and H. R. Sadeghpour, Phys. Rev. Lett. 85, 2458 (2000)

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