Ultralong-range Rydberg Molecules: Electronic Structure and Rydberg blockade

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Synopsis We describe the electronic structure and main features of ultralong-range Rydberg molecules formed by a Rydberg atom, Rb or Cs, and the diatomic molecule RbCs. We also present the first experimental observation of the Rydberg blockade due to the charge-dipole interaction between a single Rb atom and a single RbCs molecule confined in different optical tweezers.

The exotic properties of Rydberg atoms make them unique probes of their environments. In hybrid systems, they form ultralong-range molecules when combined with ground-state atoms [1, 2], ions [3], or polar molecules [4, 5], which inherit these exciting properties.

When the diatomic polar molecule is immersed into the wave function of the excited atom, the anisotropic scattering of the Rydberg electron from the permanent electric dipole moment of the dimer is responsible for the binding mechanism in these Rydberg molecules [4, 5]. In this work, we explore the electronic structure and main properties of these exotic ultralong-range molecules, which are formed by either Rubidium or Cesium Rydberg atoms interacting with RbCs. Our focus is the regime where the charge-dipole interaction of the Rydberg electron with the diatomic polar molecule induces a coupling between the quantum defect Rydberg states and the nearest degenerate hydrogenic manifold. We present adiabatic electronic states evolving from the Rydberg degenerate manifold and from the quantum defect states, and analyze the non-adiabatic coupling between these potentials, and the decay rates and formation rates of their of the bound vibrational states. We propose a protocol to create these molecules experimentally in these electronic states from a mixture of ultracold atoms and ultracold molecules [6].

In addition, we present the first experimental demonstration of the Rydberg blockade due to the charge-dipole interaction between a single Rb atom and a single RbCs molecule. The atom and molecule are confined in optical tweezers, which are used to control their relative distance. For a separation of 310(40) nm, the charge-dipole interaction between the Rydberg electron and atomic core with the dipole moment of RbCs provokes the blockade of the transition to the Rb($52s$) Rydberg state. The observed excitation dynamics are in excellent agreement with the theoretical results obtained using the electronic structure of the Rydberg Molecule Rb-RbCs [7].

References

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