

Anisotropic blockade using pendular long-range Rydberg molecules

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Synopsis Ultralong-range “butterfly” Rydberg molecules possess large dipole moments which lead to long-range anisotropic interactions between molecules. Their properties are studied using a Hamiltonian that fully includes the electronic and nuclear spin degrees of freedom. Using this more accurate and complete theoretical description, we investigate the intermolecular interaction when the molecules are prepared in pendular states. The anisotropy of this interaction manifests itself strongly in the density of created molecules.

The recent observation of “butterfly” Rydberg molecules [1] has confirmed theoretical predictions made well over a decade ago [2]. The high experimental resolution of the photoassociation spectrum, along with the additional measurements of the bond lengths and dipole moments of these molecules, necessitates a more sophisticated formulation of the theory including nuclear and electronic spin degrees of freedom to quantitatively understand this system.

We have constructed a Hamiltonian which incorporates these effects, including the dependence of the electron-atom P -wave interaction on the different 3P_J scattering phase shifts. This has been neglected in nearly all previous treatments, and our approach clarifies its derivation and demonstrates its importance [3]. This full theory has been used to calculate accurate potential curves and electronic multipole moments for both Rb and Cs, and has revealed an improved picture of recent experimental observations. We report on how this 3P_J dependence of the Hamiltonian mixes states of different molecular symmetries, reducing their dipole moments in good agreement with experiment [1].

With the improved theoretical calculations of these molecular properties in hand, we have studied the long-range intermolecular interactions. Since even weak external fields tightly align these molecules, putting them deep within the so-called “pendular” regime, butterfly molecules are ideal candidates for exploring many-body physics in dipolar gases. We have calculated the potential energy surface, dominated at long range by the anisotropic dipole-dipole interaction but also including higher-order multipole and van der Waals terms, between aligned molecules in a quasi-one-dimensional trap at an angle θ relative to the applied field. By varying θ the dipole-dipole contribution can be tuned to zero at the

magic angle $\theta_{M,n}$, which provides a clear experimental signature of this anisotropy in the density of molecules since the strong interactions “blockade” nearby excitations. This straightforward experimental scheme outlines a path towards the study of polaron/impurity physics in a mixed system, angulon/pendulon interactions, or the formation of crystal-type phases with Rydberg molecules.

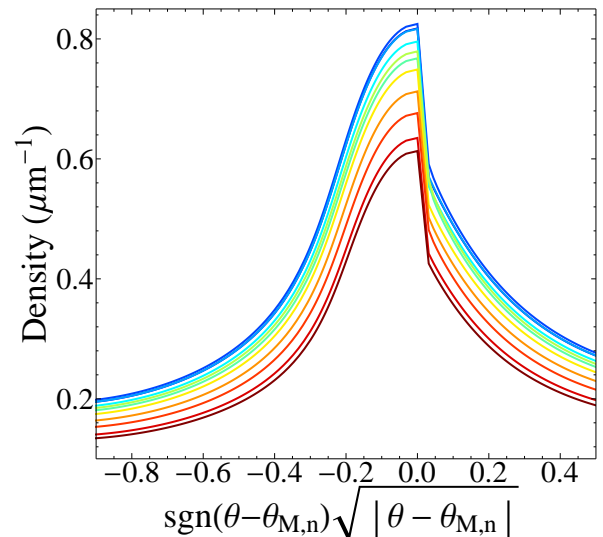


Figure 1. Simulated density as a function of the angle between the field and trap axes. As n increases from 20-40 the color changes from blue to red.

References

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