

DAMOP15-2015-000937

Abstract for an Invited Paper
for the DAMOP15 Meeting of
the American Physical Society

Photoassociation of long-range nD Rydberg molecules¹

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Cold atomic systems have opened new frontiers at the interface of atomic and molecular physics. Of particular interest are a recently discovered class of long-range, homonuclear Rydberg molecules first predicted in [1] and observed in [2]. In rubidium, these molecules are formed via low-energy electron scattering of the Rydberg electron from a $5S_{1/2}$ ground-state atom that is present within the Rydberg atom's volume. The binding mostly arises from S-wave and P-wave triplet scattering. In recent work [3], we have observed long-range homonuclear diatomic nD Rydberg molecules photoassociated out of an ultracold gas of ^{87}Rb atoms for principal quantum numbers $34 \leq n \leq 40$. Related results have also been reported in [4]. The measured ground-state binding energies of $^{87}\text{Rb}(nD + 5S_{1/2})$ molecular states are larger than those of their $^{87}\text{Rb}(nS + 5S_{1/2})$ counterparts, showing the dependence of the molecular bond on the angular momentum of the Rydberg atom. We have exhibited the transition of $^{87}\text{Rb}(nD + 5S_{1/2})$ molecules from a molecular-binding-dominant regime at low n to a fine-structure-dominant regime at high n [akin to Hund's cases (a) and (c), respectively]. In our analysis [4], we use a Fermi model that includes S-wave and P-wave singlet and triplet scattering, the fine structure coupling of the Rydberg atom and the hyperfine structure coupling of the $5S_{1/2}$ atom. The hyperfine structure is important because it gives rise to mixed singlet-triplet potentials.

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¹This work was supported by the AFOSR (FA9550-10-1-0453) and the NSF (PHY-1205559)