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Photoassociation of long-range $nD$ Rydberg molecules$^1$

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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 $^{87}$Rb($nD + 5S_{1/2}$) molecular states are larger than those of their $^{87}$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 $^{87}$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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