

Formation of rotationally-excited ultralong-range Rydberg molecules: Role of photon momentum transfer, sample temperature, and ground-state atom-atom interactions.

C. Wang, Y. Lu, S. K. Kanungo, T. C. Killian, and F. B. Dunning (Rice University)
S. Yoshida (Vienna University of Technology)

Ultralong-range Rydberg molecules (ULRRMs) comprise a Rydberg atom in whose electron cloud is embedded one, or more, ground-state atoms weakly bound through scattering of the Rydberg electron. Here we examine the factors that govern ULRRM formation, with emphasis on the production of rotationally-excited dimers. Strontium $5sns\ ^1S_0$ dimers are created in a cold gas, $\sim 1\ \mu\text{K}$, by two-photon excitation using lasers operating at 412 and 461 nm. The important role played by photon momentum transfer during excitation of one of the atoms in the initial atom pair to a high Rydberg state is examined through measurements using co- and counter-propagating laser beams. (For counter-propagating beams the net photon momentum transfer is near-zero providing a valuable benchmark against which to look for such effects.) The role of atom-atom interactions in the parent cold gas is explored through comparative studies with ^{84}Sr and ^{86}Sr which have very different s -wave scattering lengths, a_s , of 123 and 811 a_0 , respectively. For ^{86}Sr , a_s is comparable to the size of an $n\sim 25$ Rydberg atom which suppresses, for nearby values of n , the formation of dimers in the $N=0$ ground rotational state, reducing the dimer production rate and allowing the effects of higher-wave scattering, which leads to the production of rotationally-excited states, to be observed. Measurements also show that increases in sample temperature lead to increased relative production of rotationally-excited states. The results are in good agreement with the predictions of a model that includes all these factors.