

Observation of singlet-scattering channels in long-range Rydberg dimers

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Synopsis We report on the observation of long-range Cs₂ Rydberg molecules consisting of a p-state Rydberg atom and a ground-state atom. Two types of molecular states are visible in the photoassociation spectra: states bound purely by triplet *s*-wave scattering and states bound by mixed singlet-triplet *s*-wave scattering. An effective Hamiltonian including *s*-wave scattering pseudopotentials, the hyperfine interaction of the ground-state atom, and the spin-orbit interaction of the Rydberg atom reproduces the experimental observations. The detailed analysis reveals the important role of singlet scattering in long-range Rydberg molecules and yields an effective singlet *s*-wave scattering length for low-energy *e*[−]-Cs collisions.

Long-range Rydberg dimers are molecules in which one of the atoms is in the ground state and the other one is in a highly excited electronic state, a Rydberg state. The binding of these molecules is caused by the elastic scattering of the Rydberg electron off the ground-state atom, which is located within the electron orbit. This type of molecular bond was initially proposed by Greene *et al.* [1] and has been observed experimentally in the photoassociation of ultracold rubidium and cesium atoms close to transitions to *s*, *p* and *d* Rydberg states [2–7].

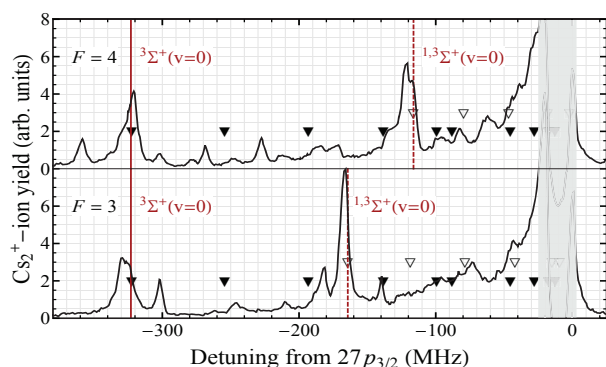


Figure 1. Detected Cs₂⁺ ions after photoassociation in an ultracold gas of cesium atoms as a function of the laser frequency near the 27p_{3/2}+6s_{1/2} (*F*=3, 4) asymptotes. The solid (dashed) red lines mark the calculated positions of the *v*=0 levels of the ³Σ⁺ (^{1,3}Σ⁺) states. Full and open triangles mark the calculated positions of higher vibrational levels of the ³Σ⁺ and ^{1,3}Σ⁺ states, respectively. The strongly saturated atomic transition corresponds to the grey area.

Here we report on the formation of long-range Rydberg molecules correlated to *np* Rydberg states of cesium with principal quantum

number *n* ranging from 26 to 34 [7]. The observed binding energies reach up to 400 MHz and the internuclear separations exceed 45 nm at *n*=26. As shown in Figure 1 for *n*=27, the photoassociation spectra reveal two types of molecular states recently predicted by Anderson *et al.* [4]: Bound states for which the binding results only from triplet *s*-wave scattering (labeled ³Σ⁺), and more weakly bound states with contributions from both singlet and triplet *s*-wave scattering (labeled ^{1,3}Σ⁺). The experimental observations are well described by a model including *s*-wave scattering, the hyperfine interaction of the ground-state atom and the fine structure of the Rydberg atom.

By adjusting the values of the zero-energy singlet and triplet *s*-wave scattering lengths to *a*_{*S*,0} = −3.5 ± 0.4 *a*₀ and *a*_{*T*,0} = −21.8 ± 0.2 *a*₀, respectively, we are able to reproduce the experimental binding energies within the experimental uncertainties. This yields the first experimental determination of the zero-energy singlet *s*-wave scattering length, which is in reasonable agreement with an extrapolation from higher-energy electron-Cs scattering data [8].

References

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