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Observation of Blueshifted Ultralong-Range Cs₂ Rydberg Molecules¹

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We observe ultralong-range blue-shifted Cs₂ molecular states near $ns_{1/2}$ Rydberg states in an optical dipole trap, where $31 \leq n \leq 34$. The accidental near degeneracy of $(n-4)l$ and ns Rydberg states for $l > 2$ in Cs, due to the small fractional ns quantum defect, leads to non-adiabatic coupling among these states, producing potential wells above the ns thresholds. Two important consequences of admixing high angular momentum states with ns states are the formation of large permanent dipole moments, $\sim 15 - 100$ Debye, and accessibility of these states via two-photon association. The observed states are in excellent agreement with theory.

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