Production of ultra-long-range Rydberg molecules in a divalent atomic system

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Synopsis
Sr₂ Rydberg molecules are created through two-photon excitation in an ultracold strontium gas contained in an optical dipole trap and are detected through atom loss from the trap. First order perturbation theory employing the Fermi pseudopotential and effective s-wave and p-wave scattering lengths is used to evaluate the binding energies of the molecular levels.

Scattering of the excited electron in a Rydberg atom from a ground state atom introduces a novel chemical bond that can bind the two together to form an ultralong-range Rydberg molecule [1]. Their creation, however, requires ultracold temperatures so that the thermal energies of the atoms are lower than their binding energies, typically ~1-100 MHz, and high-density samples to ensure a high probability for finding two atoms with separations less than the radial extent of the Rydberg electronic wave function.

Whereas several studies have reported observation of Rydberg molecules using alkali Rydberg atoms, we report here their first observation in the alkaline earths using ⁸⁴Sr atoms held in an optical dipole trap (ODT). The molecules are created by two-photon excitation via the 5s5p³P₁ intermediate state using radiation at 689 and 319 nm. After excitation, atoms remaining in the trap are released and their number determined by absorption of 461 nm light tuned to the ¹S₀-¹P₁ transition. Molecule creation is detected as trap loss and the molecular binding energies are determined by the shift of the molecular lines to the red of the atomic line.

Figure 1 shows the molecular binding energies observed using ³S₁ Rydberg states with values of n from 29 to 36 together with the results of calculations employing first-order perturbation theory. The Rydberg electron-ground state atom interaction is described using a Fermi pseudopotential and optimized effective s-wave and p-wave scattering lengths of -13.2 and 8.4 a₀, respectively. The agreement between theory and experiment is excellent and the binding energies of the most bound levels obey a 1/(n-δ)⁶ scaling law, where δ=3.371 is the ³S₁ quantum defect. The same scaling law was seen earlier in the alkalis [2]. Not all the predicted lines are observed experimentally due to difficulties in resolving closely spaced lines and the reduced excitation strengths at the lower n.

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References
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