Double Rydberg states of alkaline-earth atoms: two-electron dynamics far from the nucleus

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The motion of two electrons excited far away from the nucleus is the result of the subtle balance between their mutual Coulomb repulsion and the Coulomb attraction of the residual ionic core. In highly excited "double Rydberg" states (DRS), strong electronic correlations give rise to complex two-electron dynamics that range from chaotic motion to quasi-stable orbits depending on the relative degree of excitation of the two electrons and on the details of electronic correlation [1-3]. DRS thus represent an ideal test bed to explore the quantum mechanical three-body Coulomb problem. However, a detailed theoretical description of the energies and dynamics of DRS, combined with systematic experimental studies in the time- or frequency domains, is lacking and hampers our understanding of a seemingly simple problem: how two electrons move far away from the nucleus.

Theoretically, the recent development of the method of configuration interaction with exterior complex scaling (CI-ECS) has paved the way to accurately treating high-lying doubly excited states [2]. The method relies on a two-active-electron approach, on the use of exterior complex scaling to treat the hundreds of open channels in which DRS can autoionize, and on optimized numerical basis functions to make the CI expansion as compact as possible. In this way, the energies, lifetimes and wavefunctions of the very dense manifold of DRS resonances just below the double ionization threshold ($E\sim$ -0.5 meV) can be calculated, providing a detailed information on the associated electron correlations and dynamics [3].

In this talk, I will present the recent advances achieved with combined theoretical CI-ECS and experimental approaches in the study of the DRS of the Sr atom. I will explain (i) how such states are prepared through sequential, resonant, multiphoton excitation of the two valence electrons of Sr; (ii) how they are measured through multiple double-ionization mechanisms; and (iii) how the signatures of electronic correlations are visible in the dense and complex DRS spectra. A particularly interesting class of DRS are planetary states, in which the two-electron motion resembles the motion of two planets orbiting a star. The strong interaction between the electrons induces a dynamical localization of the electronic wavefunction (Fig. 1) and long autoionization lifetimes [1, 3]. Planetary states will be identified in the experimental spectra and their spectacular two-electron dynamics will be visualized with the calculated wavefunctions.

The results presented in this talk will provide insight into the physics of DRS and the underlying three-body problem [3], ultimately paving the way to controlling and manipulating atoms in such states at the quantum-state level and using them for, e.g., nondemolition measurements or simulation [4].

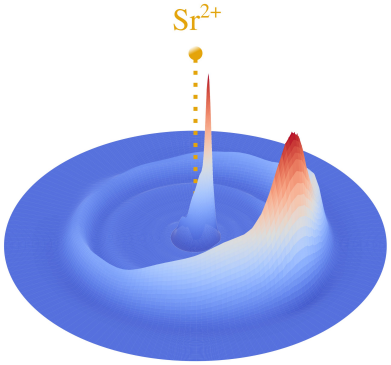

Figure 1. Two-electron composite probability density for a planetary state of the Sr atom below the N=8 ionization threshold. The composite density was obtained by summing, for each electron, the two-electron density calculated while fixing the other electron at its most probable radial distance r.

References

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