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# **EDITORIAL**

# **Cold Rydberg gases and ultra-cold plasmas**

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#### Abstract

We briefly highlight recent developments in the complementary fields of cold Rydberg gases and ultra-cold plasmas and provide a short overview of the articles in this special issue.

# 1. Introduction

The topic of Rydberg states has a long and distinguished history in atomic, molecular and optical physics [1]. The enormous transition dipoles between Rydberg states, allowing strong coupling between single atoms and photons, have been particularly fruitful in the development of cavity QED [2, 3]. Similarly, the dramatic scaling of the spatial extent of the Rydberg wavefunctions and orbital periods with principal quantum number has enabled precise control and direct imaging of the orbital motion of an electronic wave packet [4].

The advent of laser cooling and trapping techniques has opened a new domain for Rydberg atom research making it possible to realize a regime where the interaction energy between Rydberg atoms greatly exceeds their kinetic energy. This inversion of energy scales combined with precise experimental control of Rydberg states has advanced a diverse range of applications as well as fundamental science.

Barely one decade on from the pioneering experimental [5–7] and theoretical [8, 9] work on cold Rydberg systems, the field is developing at a rapid pace and continues to expand into new areas. Many of the early theoretical ideas such as fast quantum gates [8] or long range molecules [10, 11] have now been realized experimentally [12–15] and new ideas, expanding into the realm of quantum many-body physics, as well as nonlinear and quantum optics, have emerged.

This special issue provides snapshots of such ongoing developments. The contributions to this collection have been grouped into five subject areas. The first part describes the control and probing of *Rydberg atoms in external fields*, followed by investigations of *ultra-long range Rydberg molecules*. The third and fourth parts present experimental and theoretical studies of *many-body physics in cold Rydberg gases* and *ultracold plasmas*. The final part presents work on *quantum information* with Rydberg atoms, including applications to quantum optics.

Below we give a brief overview of recent developments in the context of the contributions to this special issue. For more detailed and comprehensive information we refer the reader to recent review articles on Rydberg atoms in external fields [16], Rydberg atom interactions [17, 18], quantum information [19] and ultracold plasmas [20, 21].

# 2. Rydberg atoms in external fields

Due to the simplicity of their electronic structure, most studies of cold Rydberg gases focus on alkali metals, where the effective one-electron character allows high-precision structure calculations. In the paper by Millen *et al* [22], two-electron Sr atoms are laser cooled and interrogated spectroscopically. Two-electron atoms where more than one electron is excited are prone to auto-ionization. In the experiment both spontaneous ionization, which occurs in dense Rydberg gases due to Penning ionization [23] resulting from Rydberg–Rydberg collision, and autoionization, which occurs via electron–electron correlation in double excitations in a Rydberg atom, were observed. A single-electron quantum defect model is employed to accurately calculate the transition dipole moments and compare with observation. Discrepancies

in comparison with the singlet and triplet lines in the measured Stark Rydberg maps allude to the role played by electron– electron interaction. It is shown that autoionization can be employed to state-selectively study the dynamics of such twoelectron Rydberg gases.

Dephasing of Rydberg wave packets is a subject that has found renewed interest in particular in relation to noiseinduced decoherence in cold open quantum systems [24, 25]. In the work by Wyker et al [26], Rydberg atoms in nearly circular high angular momentum states  $(l \sim n)$  and high principal quantum number  $n \sim 300$  were studied subject to stochastic electric noise and collisions. Atoms are initially prepared in zero field into high n p states, to increase the oscillator strength, and then driven into high-l states in weak electric fields. Quantum revivals of the wave packets on the time scale of hundreds of Kepler periods are observed and are suppressed in amplitude by the application of electric field noise. Collisions with the ambient gas at moderate densities are not found to be a major source of decoherence. The collisional broadening can be calculated using the Fermi pseudopotential s-wave electron-Rydberg scattering method.

In the paper by Donnan *et al* [27], the stability of deeply bound Rydberg hydrogen states in parallel constant magnetic and time-increasing electric fields is investigated. These calculations are useful for understanding the dynamics of atom formation in antihydrogen traps. Whereas, in alkali-metal atoms, selective-field ionization due to time-increasing electric field can occur at low angular momentum states at the classical ionization threshold (because of the large quantum defects of such states), in hydrogen atoms, the pure Coulomb potential allows stable states to form above the classical ionization threshold. In a combined electric and magnetic field, the classical hydrogen atom is chaotic and so the adiabaticity of level crossings becomes an issue. Both classical and quantum methods are used to calculate the survival probability of the H(n = 30 and 60) state in magnetic field of a 4 tesla field and varying electric fields. The calculations confirm the appearance of large field peaks indicative of classical orbits which survive ionization above the ionization threshold. Delayed ionization has implications for the recombination of weakly bound states in anti-hydrogen traps.

#### 3. Ultra-long range Rydberg molecules

Owing to their enormous polarizability Rydberg atoms can facilitate the formation of exotic molecules. One such molecule, comprised of a ground state atom, bound at large distances (thousands of Bohr radii) within the electronic wavefunction of another Rydberg atom, forms via frequent electron–atom collisions [10, 14, 28–36], while the strong long-range interaction between two Rydberg atoms can give rise to Rydberg macrodimers with even larger bond lengths [11, 15, 37–41].

Understanding the mechanisms leading to the decay of these Rydberg molecules is important for future experiments and possible applications [42]. This question is addressed in the paper by Butscher *et al* [43] for Rb(5 s-n s) ultra-long range molecules. The lifetime is shown to be systematically

shorter than the lifetime of bare atomic Rydberg states and depend on the temperature and density of the gas as well as on the particular vibrational state. The experimental findings are explained within a simple binary collision model and by including an additional decay mechanism [36] arising from the inward penetration of bound atomic pairs due to imperfect quantum reflection, which would otherwise stabilize excited molecular states.

An even more exotic variant of such long-range molecules is possible when the ground state atom is replaced by a polar molecule. The interaction between the Rydberg atom and the polar molecule can no longer be represented in the Fermi short-range pseudopotential, as the electron-dipole scattering does not have a scattering length and is a very long-range  $1/r^2$  interaction. In the paper by Rittenhouse *et al* [44], the technique developed in [45] for a degenerate Rydberg manifold is refined further to include nearby (n + 3) s Rydberg states in rubidium atoms. It is shown that the non-adiabatic interactions between the s and the zero-quantum defect degenerate Rydberg manifold can be 'tuned' with the size of the polar molecule permanent dipole moment. The admixture of s character into the degenerate high-angular momentum states opens up the possibility of formation of such controllable molecules with standard two-photon photoassociation techniques.

The paper by Samboy and Côté [46] investigates longrange bound macrodimers resulting from the strong interaction between two distant rubidium Rydberg atoms. Using symmetry-adapted atomic Rydberg basis states, the asymptotic molecular Hamiltonian is diagonalized [41], and long-range potential wells, with a depth of several GHz and large equilibrium distances between  $10^4$  and  $10^5$  Bohr radii, are found. The paper derives simple scaling relations between the potential properties and the atoms principal quantum number, and presents detailed calculations on a possible scheme to produce such molecules in an optical lattice by photoassociation.

The effect of an electric field onto the long-range interactions between Rydberg atoms is the subject of the experimental/theoretical study presented in the paper by Cabral *et al* [47]. Born–Oppenheimer potential energy curves for Rb and Cs Rydberg atoms at large distances were calculated in a constant electric field. A nontrivial dependence on the atomic species is pointed out, which precludes simple extrapolation and necessitates explicit calculations for each species. The rate constant for the production of Rydberg atoms is calculated using a density matrix formalism and good agreement with the experimental observations is found.

## 4. Many-body physics using Rydberg atoms

In 2004, two independent experiments provided clear evidence of the profound effects of strong van der Waals interactions in cold Rydberg gases by measuring a dramatic suppression of Rydberg excitation due to interaction-induced level shifts in cold rubidium gases [48, 49]. Soon after this first observation of the *Rydberg blockade* numerous experimental and theoretical works appeared investigating the underlying manybody excitation dynamics and the resulting strong correlations

Several experimental schemes between Rydberg states. [50–55] have characterized the Rydberg blockade in frozen gases for different types of interactions, angular momentum states and atomic species [56-64, 66, 67], and different theoretical approaches have been developed to describe and understand the experimental observations [48, 68–78, 110]. More recently, theoretical work has established a fruitful analogy between laser-driven Rydberg gases and long-range interacting spin systems, elucidating the many-body excitation dynamics in terms of spin relaxation or thermalization processes [79–81] or in terms of adiabatic parameter changes within the ground state phase diagram of the corresponding many-body spin Hamiltonian [82-85]. The latter notion, for instance, has led to the prediction of long-range ordered, crystalline Rydberg atom structures [82–84], whose properties and stability are currently investigated by several groups [86–89].

In [90] of this special issue, van Bijnen *et al* present a detailed study of such Rydberg crystals and analyse their creation from a frozen gas of cold atoms via excitation by chirped laser pulses. For the particular case of cold and ultracold rubidium gases, the authors discuss the experimental feasibility and limitations of such a scheme and outline a corresponding experiment, set up by the group at Eindhoven University. Assuming a perfectly adiabatic preparation, a broad range of crystalline structures in one, two and three dimensions is presented. In one dimension, the dynamics for various trap geometries, such as optical lattices, dense Bose– Einstein condensates and thermal cloud in a optical dipole trap or magneto-optical trap, is considered. This work identifies optimal excitation schemes for each of these situations, which will be important for future experiments.

The paper by Tezak *et al* [91] investigates the many-body energy spectrum of repulsively interacting Rydberg atoms in a one-dimensional lattice [92]. The generally complicated energy spectrum is approached from two limiting sides: (i) the low-intensity limit where the spectral properties are dominated by the strong Rydberg–Rydberg atom interactions, and leads to a sequences of crystal states, and (ii) the strong driving limit, which can be perturbatively described in terms of fermionic excitations with small energy corrections due to the van der Waals interactions. The in-depth theoretical discussion, stretching between these two limiting cases, illustrates the rich the behaviour of laser-driven Rydberg lattices and provides valuable insights for further work to characterize the response of cold Rydberg gases to different excitation schemes [93].

The formation of an optical lattice of alkaline earth, e.g. Sr, Rydberg atoms [94, 95], is described in the paper by Mukherjee *et al* [96]. The proposed approach exploits the second-valence electron available in alkaline–earth atoms, whose strong polarizability is shown to enable strong optical Rydberg atom traps, providing an alternative mechanism to ponderomotive level shifts used in alkaline atoms [97–100]. This work identifies magic wavelengths, in an accessible frequency range, that permit strong simultaneous lattice confinement of ground state atoms and Rydberg atoms in the same trapping potential. In contrast to *n*S Rydberg states of alkaline atoms [101–103], the presented calculations reveal attractive van der Waals interactions between  $Sr(^1S_0)$  atoms, which are shown to yield a many-body energy spectrum that dramatically differs from the repulsive case described above and may permit the creation of highly entangled many-body GHZ (Greenberger–Horne–Zeilinger) states via chirped-pulse excitation.

Besides strong van der Waals interactions, Rydberg atoms also exhibit strong dipole-dipole couplings between states with adjacent angular momenta. This coupling becomes important in Rydberg gases with mixed excitations [5, 65, 104, 105] or in the presence of Förster resonances [6, 66, 127]. Experiments and theoretical investigations on excitation transfer in cold Rydberg gases [106–111] show that strong dipole-dipole interactions lead to resonant migration of Rydberg excitations through the ensemble on very fast timescales much shorter than transport times for motional diffusion. The coherence properties of dipolar excitation transfer in cold Rydberg systems have been investigated for two small localized clouds of atoms [65, 112-114] and, theoretically, in atomic chains, proposing a cold-atom realization of quantum random walks and exciton transport models [115, 116].

Exciton transport is studied in the paper by Möbius et al [117] where the dynamics of a nP Rydberg excitation via dipole-dipole coupling to surrounding nS states is investigated in a flexible Rydberg atom chain [118-120]. It is pointed out that the strong dipole-dipole interaction not only facilitates fast transport of the p excitation but also leads to motion of the Rydberg atoms, which is shown to strongly entangle the motional degrees of freedom and the internal many-body state of the Rydberg atom chain. For proper initialization of the system, it is demonstrated that entanglement can be locally transported alongside the p excitation, illustrating the intricate interplay between excitonic and translational motion. The results demonstrate excellent agreement between a full quantum calculation for small systems (of the internal and translational dynamics) and a mixed quantum-classical treatment [121], which facilitates the description of much larger systems and possibly more complex scenarios of exciton transport.

The paper by Scholak *et al* [122] investigates excitation transport via dipolar coupling through a random arrangement of particles [123, 124], with particular emphasis on the competing influence of coherence and de-phasing onto the transport efficiency of such a quantum network. Defining different measures to characterize the transport efficiency, the authors identify optimal spatial configurations. Interestingly, these optimal configurations maximize the transport efficiency for purely coherent dynamics, in marked contrast to the majority of networks, for which transport is typically enhanced by environmental noise.

The investigations by Möbius *et al* and Scholak *et al* are of relevance to other exciton systems, such as molecular aggregates, including biological light-harvesting complexes or organic solar cells [125]. As pointed out in both works, the close analogy between *Rydberg aggregates* and these more complex systems suggests that cold Rydberg atoms could provide an experimentally well accessible laboratory to

elucidate the basic mechanisms of energy transport by offering high-level control of the network topology, dipolar coupling strengths as well as external sources of noise and decoherence.

# 5. Ultracold plasmas

At very a high density of Rydberg atoms, their strong dipole– dipole interaction can also cause autoionization of Rydberg atom pairs [126–128] and initiate an avalanche-like conversion of a cold Rydberg gas into an ultracold plasma [129–131] signifying their deep connection as two different metastable states of one and the same system [132]. Since the first direct production of an ultracold neutral plasma via photoionization of laser-cooled atoms [7] these systems have proved to provide an appealing and well controllable platform to investigate a diverse range of plasma physics phenomena, including, e.g., plasma expansion into vacuum [133–137], recombination of Rydberg atoms [138–142], plasma instabilities [143, 144] and collective waves [145–148].

Collective ion acoustic modes are studied in the paper McQuillen *et al* [149] via fluorescence imaging of an ultracold strontium plasma. The authors demonstrate a novel approach to inscribe small-wavelength periodic modulations onto the initial plasma density, which at the lowest achieved wavelengths will enable detailed experimental studies of electron correlation effects on the character of collective ion oscillations. The described high-resolution density-sculpting technique could also permit more stringent tests of existing expansion models for ultracold plasmas and open a range of intriguing perspectives, including, e.g., investigations of the formation and dynamics of shock waves in ultracold neutral plasmas.

Owing to their low temperature, ultracold plasmas occupy an exotic parameter regime of plasma physics in which the interaction between ions can greatly exceed their thermal energy. As a striking consequence of the resulting correlations, experiments observed strong ion heating [150] characterized by pronounced temporal temperature oscillations [151, 152]. The paper Lyon and Bergeson [153] reports a detailed study of electron screening effects on the corresponding temperature evolution during the early stages of the plasma evolution. The presented fluorescence measurements of an ultracold calcium plasma reveal a significant influence of electron screening on the observed correlation-induced ion heating, which is found to be larger than observed previously.

Recently, a new type of ultracold plasma has emerged, produced via Rydberg excitation of cold molecules in a supersonic molecular beam [154, 155]. Due to their substantially larger density and the possibility of ultracold chemical reactions involving highly excited Rydberg molecules, these plasmas are expected to behave differently from the atomic plasmas discussed above, and have, consequently, raised a number of interesting questions. In [156] Saquet *et al* describe such an experiment, in which an ultracold plasma is created from a seeded supersonic beam of nitric oxide molecules. The presented time-offlight measurements reveal an expansion behaviour that is well described within a hydrodynamic plasma model, while suggesting an electron temperature that is substantially lower than that found in ultracold atomic plasmas. As a first step towards understanding this surprising result, the paper presents kinetic model calculations to track the collisiondriven time evolution of the Rydberg molecule population in the plasma. This work also points out a number of important questions, such as strong coupling effects and the role of low-temperature chemical reactions, that will need to be addressed towards a comprehensive understanding of the complex plasma dynamics.

#### 6. Quantum information

A comprehensive review of quantum information using Rydberg atoms has recently appeared [19]. Consequently, here we will only focus on recent developments relevant to the papers in this special issue. One can distinguish between four main areas where Rydberg atoms exhibit useful properties for quantum information applications. The first is in cavity QED where the large dipole moment associated with microwave transitions between Rydberg states enables the strong coupling regime between single photons and single atoms to be achieved [2, 3]. This area is distinct from the main topic of this special issue in that experiments to date focus on one atom at one time in the interaction region and do not involve direct atom–atom interactions.

The other three areas all exploit the strong dipole– dipole interactions between atoms, i.e. the dipole blockade mechanism. First there are blockade effects where the atoms are individually addressable [12, 13, 157–164], second blockaded ensembles [9, 165–176] and third applications in quantum optics where the blockade effect is exploited to modify a light field [177–187]. Of the five special issue papers in these categories, the first (Møelmer *et al* [188]) deals with the implementation of Grover's algorithm in a Rydberg ensemble, and the other four [189–192] discuss light-matter interactions in Rydberg ensembles.

#### 6.1. Quantum algorithms in Rydberg ensembles

One attractive feature of the Rydberg blockade mechanism is that a single Rydberg excitation can control the behaviour of  $N_b$  atoms its vicinity, where  $N_b$ , the number of atoms in the blockade sphere, can be up to of order 1000 [60]. This property allows quantum information protocols to exploit multi-particle entangled states [168, 170] and mesoscopic quantum gates [171].

The paper Mølmer *et al* [188] takes this idea a stage farther by proposing a protocol to implement Grover's search algorithm using the Rydberg blockade mechanism. This approach exploits the multi-atom approach inherent to Rydberg systems as opposed to the more standard two-qubit circuit model, and thereby could offer considerable practical advantages, particularly with regard to the scaling of errors.

## 6.2. Quantum and nonlinear optics using Rydberg ensembles

Rydberg ensembles or individual Rydberg superatoms offer significant adavantages for single-photon nonlinear optics due

to their large optical cross section. The cross section is enhanced by the number of atoms per blockade sphere  $N_{\rm b}$ which as discussed above can be of order of 1000. In addition, as a superatom is larger than an optical wavelength, the optical emission properties can be highly directional [193, 194]. For this reason, a Rydberg ensemble is a promising candidate for a deterministic and directional single photon source [193, 194]. The paper by Laycock et al [189] considers a particular case of single-photon emission from a 2D lattice of emitters prepared in many-body states using van der Waals interactions between highly excited Rydberg states. The protocol prepares single photons in highly exotic spatial modes which in addition to their intrinsic interest could be of use in quantum networks.

A second rapidly developing area is electromagnetically induced transparency (EIT) [195] involving highly excited Rydberg states [196]. Recently, it has been shown that the combination of EIT and strong dipole–dipole interactions between highly excited Rydberg atoms gives rise to giant 'Kerr-like' optical nonlinearity [178, 185], and the formation of non-classical states of light [184]. The large single-photon nonlinearities accessible in Rydberg systems enable strong photon–photon interactions and the possibility of photonic phase gates [177, 181, 183].

In this special issue there are three papers which cover different aspects of EIT and coherent population trapping (CPT) [197] involving highly excited Rydberg states. The paper by Sevinçli et al [190] combines EIT and CPT [197] and highlights the universal scaling of the excitation in threelevel Rydberg ensembles [182]. The paper by Pritchard et al [191] investigates the transient nature of the optical nonlinearity due to motional effects in the Rydberg gas. The paper considers the case of Rydberg atoms with attractive dipole-dipole interactions where the motional effects are more pronounced than for repulsive interactions [178]. The paper by Tanasittikosol et al [192] shows that a dramatic enhancement of the optical nonlinearity can be achieved by dressing the Rydberg state with a resonant microwave field. The enhancement occurs due to an effective increase in the number of atoms per blockade sphere. The ability of microwave fields to modify the interactions between Rydberg atoms provides a very powerful tool for applications in quantum information and quantum optics.

## 7. Summary

As illustrated in this special issue, Rydberg atoms combine several intriguing properties—*extreme controllability* by external fields, *strong long-range interactions* and *long lifetimes*—that make them a near-perfect system with which to explore and exploit few- and many-body quantum phenomena. At the same time, they provide a paradigm for quantumclassical correspondence and a route to strongly correlated classical plasmas. The broad range of topics covered in this special issue reflects the diversity of research that has emerged from these perspectives, and further directions may well be expected to arise considering the current pace of new developments. While this collection of articles can only be a compendium of current activities, we hope that it will provide an inspiring reference for the community and serve as a useful resource for scientists entering the field.

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