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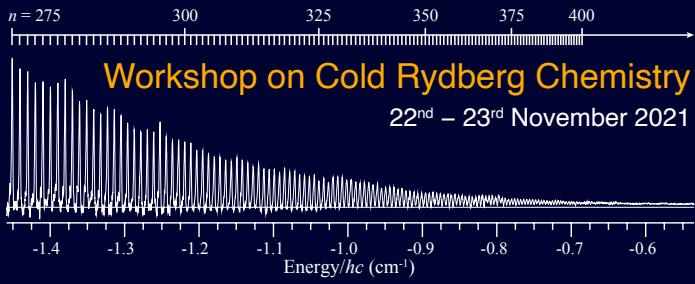
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## Timetable

Monday 22<sup>nd</sup> November 2021

Time (GMT)	
14:00	<b>Introduction</b>
<b>Collisions</b>	Chair: <i>Hossein Sadeghpour</i>
14:05	Using electron collision methods to characterise Rydberg states <i>Jonathan Tennyson</i>
14:35	Towards detecting quantum-Langevin behavior in ion-molecule reactions <i>Frédéric Merkt</i>
15:05	Self-organization in the avalanche, quench and dissipation of a molecular ultracold plasma <i>Edward Grant</i>
15:35	Cold Rydberg-ion collisions <i>Jesus Pérez-Ríos</i>
16:05	<b>Break</b>
<b>Hot Topic Talks</b>	Chair: <i>Stephen Hogan</i>
16:15	Studying ion-molecule reactions within the orbit of a Rydberg electron – the effect of the $\Lambda$ -doubling in NO on the $\text{He}^+ + \text{NO}$ reaction rate coefficient in the $k_B \cdot (0 - 10)$ K collision-energy range <i>Valentina Zhelyazkova</i>
16:22	Electrically-tuned Förster resonances in cold collisions of Rydberg-stated-selected helium atoms with ground-state ammonia molecules <i>Junwen Zou</i>
16:29	Towards quantum simulation using Rydberg states of multi-electron atoms: First spectroscopy results on erbium <i>Manfred Mark</i>
16:36	Synthetic dimension-induced conical intersections in Rydberg molecules <i>Matthew Eiles</i>

<b>Time (GMT)</b>	
<b>Atom-Ion Bound States</b>	Chair: <i>Stephen Hogan</i>
18:00	Rydberg-atom-ion molecules: Vibrational levels and lifetimes <i>Georg Raithel</i>
18:30	Search for novel atom-ion Rydberg molecules <i>Johannes Hecker Denschlag</i>
19:00	Spatial imaging of a novel type of molecular ions <i>Tilman Pfau</i>
19:30	Forming Rydberg molecules in a quantum gas with strong interactions: Rotational structure, correlations, and surprises <i>Tomas Killian</i>

## Tuesday 23<sup>rd</sup> November 2021

<b>Time (GMT)</b>	
<b>Long-Range Rydberg Molecules</b>	Chair: <i>James Shaffer</i>
14:00	How many ground state atoms can a Rydberg atom accommodate? <i>Jan-Michael Rost</i>
14:30	The life and fate of ultra-long-range Rydberg molecules <i>Herwig Ott</i>
15:00	Exploring the binding of long-range Rydberg molecules <i>Michael Peper</i>
15:30	Polyatomic ultralong range Rydberg molecules <i>Rosario González-Férez</i>
16:00	Double Rydberg molecules <i>Thomas Gallagher</i>
<b>Poster Session</b>	
16:30 – 17:15	

<b>Spectroscopy and Dynamics</b>	Chair: <i>Johannes Deiglmayr</i>
18:00	Spectroscopy and dynamics of an ion within a Rydberg-electron orbit <i>Matthieu Gévelor</i>
18:30	Quantum defects for molecular Rydberg states and precision spectroscopy of H <sub>2</sub> <sup>+</sup> <i>Maximilian Beyer</i>
19:00	Vibrational autoionization of core-nonpenetrating Rydberg states of NO: Long-range <i>ab initio</i> model and applications to cold ion chemistry <i>Timothy Barnum</i>
19:30	Effects of weak rotational and vibrational interactions on the lifetimes of long-lived Rydberg states of NO <i>Matthew Rayment</i>
20:00	<b>Closing Remarks</b>

**Part I**

**Talks**

## Using electron collision methods to characterise Rydberg states

**Jonathan Tennyson**

Department of Physics and Astronomy, University College London, UK

The R-matrix method is widely used to study a range of electron collision problems. However use scattering a negative energies has been found to be a much more effective method of characterising Rydberg states than traditional quantum chemistry techniques, which struggle reproduce high  $n$  states with conventional basis sets. The utility of the scattering method for obtain potential energy curves of many Rydberg states will be demonstrated by calculations on the nitrogen molecule.

## Towards detecting quantum-Langevin behavior in ion-molecule reactions

**K. Höveler<sup>1</sup>, J. Deiglmayr<sup>1,2</sup> and F. Merkt<sup>1</sup>**

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At very low temperatures or collision energies, the rates of ion-molecule capture reactions involving polarizable, nonpolar molecules have been predicted theoretically to increase by a factor of two compared to the Langevin rate [1-4]. This purely quantum-mechanical effect has not been observed experimentally yet and its observation represents one of the great challenges of cold ion-molecule chemistry. The study of ion-molecule reactions at low collision energies ( $E_{\text{coll}}$ ) below  $E_{\text{coll}}/k_{\text{B}} = 10$  K, or low temperatures, is experimentally very difficult because stray electric fields in the reaction volume heat up the ion samples. A potential difference of 1 mV across a reaction region of 1 cm accelerates the ions to 1 meV, which corresponds to heating them up to about 12 K. To overcome this problem and study ion-molecule reactions below 10 K, we have developed a new method, in which the ion-molecule reaction takes place within the orbit of a Rydberg electron at high values of the principal quantum number  $n$  [5]. In high- $n$  Rydberg states, the Rydberg electron only very weakly interacts with the ion core, so that it does not significantly influence the ion-molecule reaction taking place within its orbit but shields the ion from heating by stray electric fields. Instead of studying exothermic and barrier-free ion-molecule reactions of the type



we study the reactions



in which  $A^*$  and  $C^*$  represent atoms or molecules in high Rydberg states with ion cores  $C^+$  and  $D^+$ , respectively.



To reach very low collision energies, we use chip-based Rydberg-Stark decelerators and deflectors [6,7] to merge cold supersonic beams of  $A^*$  and  $B$  and to vary their relative velocities [5]. By monitoring the yield of the products ( $C^*$ , or  $C^+$  after autoionization of  $C^*$ ) as a function of the relative mean velocity of the two beams, we obtain the relative reaction cross sections as a function of the collision energy. At collision energies ( $E_{\text{coll}}/k_B$ ) below 1 K, we find that the reaction rate coefficients deviate from those estimated with Langevin-type capture models [8-12]. The deviations become particularly large when  $B$  has a permanent dipole [9,10] or a quadrupole moment [12]. To detect the factor-of-two enhancement predicted for quantum-capture reactions, it is advantageous to study reactions of ions with neutral molecules that do not have a quadrupole moment. The talk will present recent results of studies of the reactions of ground-state  $X^+ \ ^2\Sigma_g^+(v^+ = 0, N^+ = 0)$   $H_2^+$  ions with ground-state  $X \ ^1\Sigma_g^+(v = 0, N = 0)$  HD and para- $H_2$  molecules which reveal first signatures of quantum capture in ion-molecule reactions.

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## Self-organization in the avalanche, quench and dissipation of a molecular ultracold plasma

**Ruoxi Wang<sup>1</sup>, Kevin Marroquin<sup>2</sup>, John Sous<sup>3</sup>, Abhinav Prem<sup>4</sup>, James Keller<sup>2,5</sup> and Edward Grant<sup>1,2</sup>**

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Strong correlation operates in many-body systems out of equilibrium to constrain the mobility of matter and energy. Local potentials oppose ergodic driving forces and trap systems in local minima. Diverse emergent phenomena, including glassiness, topological phases, quantum magnetism, fractional quantum Hall states, and high-temperature superconductivity, all owe defining characteristics to constrained or directed transport either in the classical or quantum regime. Rydberg gases offer particular advantages as materials from which to form out-of-equilibrium strongly correlated many-body systems. Rydberg-Rydberg interactions occur with exaggerated coupling energies in systems of rarified density. Atomic Rydberg ensembles exhibit cooperative properties that range from precisely defined pairwise and higher-order coherent phenomena to aggregation, dissipation, non-equilibrium phase transitions and avalanche to plasma. Experiments elsewhere have established evidence for self-organizing criticality in ultracold and room-temperature atomic gases. Here, we explore the dynamical properties of the out-of-equilibrium arrested state produced when a molecular Rydberg gas of nitric oxide entrained at milli-Kelvin temperature in a skimmed supersonic beam evolves via a sequence of electron-impact avalanche, bifurcation and quench to form a strongly coupled, ultracold plasma. This system naturally populates energy bands with an order determined by constraints that link molecular predissociation rate to Rydberg orbital angular momentum. Inter-band Rydberg-Rydberg transitions exhibit

Fano lineshapes that reflect the degree to which bright states interact with a network of coupled Rydberg molecules that forms a spatially correlated system of randomly interacting dipoles of random energies. Particular spectroscopic transitions to a subset of highly dissociative states between bands drive the ensemble to form an open quantum system, with striking consequences evident in the collapse and dissipation of the quenched system. A description of these dynamics must account for a very large number of Rydberg and excitonic states linked by this unconventional web of many-body interactions. This condition vastly exceeds the complexity of quantum optics in a conventional few-level scheme. We have taken steps to model this system in the limit of a semi-classical many-body spin model including dissipation and disorder.

INVITED

## Cold Rydberg-ion collisions

**Jesus Pérez-Ríos**

Department of Physics and Astronomy, Stony Brook University, Stony Brook, NY 11794, USA

Rydberg-ion collisions present a strong long-range interaction that, in principle, translates into a significant charge-exchange cross section that scales quadratically with the size of the Rydberg orbit. This talk explores this reaction by identifying the different dynamical regimes regarding collision energy, finding that charge exchange and  $\ell$ -mixing collisions have complex behavior. In particular, the expected geometric scaling of the cross-section is only valid at high collision energies, whereas, in cold environments, we find significant deviations from it and that the core-electron interaction potential and core repulsion play an essential role. Our findings help to understand better the effects of Rydberg-ion long-range interaction in cold chemistry experiments.

Studying ion-molecule reactions within the orbit of a Rydberg electron  
– the effect of the  $\Lambda$ -doubling in NO on the  $\text{He}^+ + \text{NO}$  reaction rate  
coefficient in the  $k_B \cdot (0 - 10)$  K collision-energy range

Valentina Zhelyazkova, **Serena Schilling**, **Fernanda B. V. Martins**,  
and **Frédéric Merkt**

ETH Zurich, Laboratory of Physical Chemistry, Zurich 8093, Switzerland

Fast, exothermic and barrierless ion-molecule reactions drive rich chemistry in the cold ( $\sim 10 - 150$  K) and tenuous environment of the interstellar medium [1, 2]. These reactions proceed with high rate coefficients even at low temperatures, and are typically modelled by the classical Langevin model, which predicts a temperature- and collision-energy-independent reaction rate coefficient ( $k_L$ ). At low temperatures and collision energies ( $E_{\text{coll}}$ ), however, the rotational-state-dependent Stark shifts experienced by the molecule in the electric field of the ion can lead to a strong modification of the interaction potential and reaction rate coefficients [3, 4]. These Stark shifts arise from the presence of molecular dipole and, to a lesser extent, quadrupole moments and can lead to the reaction being suppressed in some molecular states or proceeding with enhanced rate coefficient in other states at the lowest collision energies [5-7].

We present experimental data and calculations on the reaction between  $\text{He}^+$  and nitric oxide in the collision-energy range  $k_B \cdot (0 - 10)$  K. In order to suppress heating by stray electric fields, the  $\text{He}^+$  ions are replaced by helium atoms in a Rydberg state [ $\text{He}(n)$ ]. To reach low collision energies, we employ the merged-beam approach [6,8]. The  $\text{He}(n)$  atoms are merged with a supersonic molecular beam containing NO molecules using a curved Rydberg-Stark surface-electrode deflector [9, 6]. The energy of the collision is varied by changing the velocity of the  $\text{He}(n)$  atoms with the deflector, while the velocity of the NO beam is kept fixed. The reaction product ions are collected in a time-of-flight mass spectrometer.

We observe a strong enhancement of the total reaction yield with decreasing collision energy, and a particularly sharp increase below  $\sim k_B \cdot 1$  K, with the total capture rate coefficient reaching values of  $\sim 3k_L$  at the lowest energies ( $\sim k_B \cdot 100$  mK). This increase is attributed to the dipole moment of NO ( $\mu_e^{\text{NO}} = 0.159$  D), the effect of which

is strongly enhanced by the presence of the  $\Lambda$ -doubling (NO has a  $X^2\Pi_{1/2,3/2}$  electronic ground state). To emphasise this effect, we also present results on the  $\text{He}^+ + \text{CO}$  reaction. Carbon monoxide has a similar dipole moment to nitric oxide ( $\mu_e^{\text{CO}} = 0.112 \text{ D}$ ), but it has a  $X^1\Sigma^+$  ground electronic state and thus no  $\Lambda$ -doubling. The total product ion yield of the  $\text{He}^+ + \text{CO}$  reaction exhibits a *decrease* with decreasing  $E_{\text{coll}}$  [7]. This effect is attributed to the negative quadrupole moment of CO ( $Q_{zz} = -9.470 \times 10^{-40} \text{ C m}^2$ ).

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# Electrically-tuned Förster resonances in cold collisions of Rydberg-stated-selected helium atoms with ground-state ammonia molecules

**Junwen Zou and Stephen Hogan**

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Collisions at low translational temperatures between atoms in Rydberg states with large electric dipole transition moments, and polar ground-state molecules represent an ideal setting to study Förster resonance energy transfer between electronic and nuclear degrees of freedom [1-4]. This type of atom-molecule interaction has applications, for example, in quantum sensing and coherent control [5-7]. Here we report intrabeam collision experiments with ground-state NH<sub>3</sub> molecules, and He atoms in triplet Rydberg states with principal quantum numbers,  $n$ , between 38 and 43. The pulsed supersonic beams used in this work were formed of NH<sub>3</sub> seeded in He at a ratio of 1:99. This resulted in a relative speed of the atoms and molecules in the moving frame of reference of  $\sim 70$  m/s, and hence relative collision energies approaching  $E_{\text{coll}}/k_B \sim 1$  K. Electric fields of up to 8 V/cm were used to tune selected Rydberg-Rydberg transitions into resonance with the NH<sub>3</sub> inversion intervals. At the collision energies accessible in this work, the resonant energy transfer observed was generally dominated by dipole-dipole interactions between the atoms and molecules. The Förster resonance widths have been found to be strongly Rydberg-state dependent. This has been inferred by comparison of the experimental data with the results of numerical calculations of the Stark shifts, and electric-field-dependent transition dipole moments in the Rydberg states.

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## Towards quantum simulation using Rydberg states of multi-electron atoms: First spectroscopy results on erbium

**Manfred Mark**

Center for Ultracold Atoms and Quantum Gases, University of Innsbruck, Austria

We present the first step towards the buildup of a versatile quantum simulator using Rydberg states of the multi-electron atom erbium in optical tweezers. Our experimental and theoretical study comprises the Rydberg spectrum for series connected to the two lowest  $4f^{12}(3H_6)6s$ ,  $J_c = 13/2$  and  $J_c = 11/2$  ionic core states of erbium. We use an all-optical detection scheme based on electromagnetically induced transparency in an atomic beam and identify approximately 550 states. Calculations using a multi-channel quantum defect theory (MQDT) shows good agreement and allows us to assign most of the states to the  $ns$  or  $nd$  Rydberg series. We also identify a state presumably from the  $ng$  Rydberg series, which we are able to couple to due to the special open-shell structure of erbium. We provide an improved accuracy for the lowest two ionization thresholds and the corresponding quantum defects for all observed series. Our results path the way for the buildup of a unique quantum simulator with erbium Rydberg states, exploiting specific lanthanide features in combination with a flexible optical tweezer setup.



# Synthetic dimension-induced conical intersections in Rydberg molecules

**Matthew Eiles**

Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

We observe a series of conical intersections in the potential energy curves governing the collision between a Rydberg atom and a ground-state atom [1]. This apparent contradiction of the von Neumann-Wigner theorem is resolved by employing the energy of the Rydberg electron as a synthetic dimension. One observable consequence of the conical intersections arises in the  $\ell$ -changing collision rate of the process  $\text{Rb}(nf)+\text{Rb}(5s)\rightarrow\text{Rb}(n\ell > 3)+\text{Rb}(5s)$ . In the vicinity of a conical intersection, this rate is strongly suppressed. We expect conical intersections and other beyond Born-Oppenheimer effects to play an important role in the dynamics of Rydberg-neutral collisions and Rydberg molecules.

## References

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## Rydberg-atom–ion molecules: Vibrational levels and lifetimes

**Georg Raithel**

Physics Department, University of Michigan, USA

We present calculations on a novel type of Rydberg dimer consisting of a Rydberg-state atom bound to an ion. The molecule is formed through long-range electric-multipole interaction between the Rydberg atom and a point-like ion. Potential energy curves that are asymptotically connected with Rydberg  $nP$ -states of rubidium or cesium are found to be conducive to metastable Rydberg-atom-ion molecules with many bound vibrational states. We compute vibrational spectra, and obtain non-adiabatic-decay rates using the Born-Huang representation. The decay rates generally increase with Rydberg-state principal quantum number and vibrational quantum number. Irregularities in the decay rates are caused by interference of multiple decay paths on the potential energy curves.

## Search for novel atom-ion Rydberg molecules

**Johannes Hecker Denschlag**

Institute of Quantum Matter, University of Ulm, Germany

Recently our group has predicted a novel ionic Rydberg molecule, which consists of a neutral Rydberg atom and an atomic ion at large internuclear distance [1]. The binding mechanism is based on a sign flip in the polarizability of the Rydberg atom in the electrical field of an ion at a particular distance. Besides explaining how this atom-ion Rydberg molecules works and what its properties are, I will talk about our search for it and the difficulties that we met in our ion-trap setup.

### **References**

[1] 'Long-range atom-ion Rydberg molecule: A novel molecular binding mechanism', Markus Deiß, Shinsuke Haze, and Johannes Hecker Denschlag, *Atoms* **9**, 34 (2021).

## Spatial imaging of a novel type of molecular ions

**Tilman Pfau**

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Atoms with a highly excited electron, called Rydberg atoms, can form unusual types of molecular bonds. The bond differs from the well known ionic and covalent bonds not only by its binding mechanism, but also by its bond length ranging up to several micrometres. Here, we observe a molecular ion based on the interaction between the ionic charge and a flipping induced dipole of a Rydberg atom with a bond length of several micrometres that was recently proposed by the Raithel and the Hecker-Denschlag group. We measure the vibrational spectrum and spatially resolve the bond length and the angular alignment of the molecule using a high-resolution ion microscope. As a consequence of the large bond length, the molecular dynamics is extremely slow. These results pave the way for future studies of spatio-temporal effects in molecular dynamics, e.g., beyond Born-Oppenheimer physics.

## Forming Rydberg molecules in a quantum gas with strong interactions: Rotational structure, correlations, and surprises

**T. C. Killian<sup>1</sup>, Yi Lu<sup>1</sup>, J. Whalen<sup>1</sup>, S. K. Kanungo<sup>1</sup>, F. B. Dunning<sup>1</sup>, S. Yoshida<sup>2</sup> and  
J. Burgdorfer<sup>2</sup>**

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Ultralong-range Rydberg molecules (ULRRMs), formed from the binding of one or more ground-state atoms within the electron cloud of a Rydberg atom, are of significant interest because of their novel physical properties and their potential as probes of many-body states of quantum gases. In this talk, I will report on the creation of ULRRMs through photo-association in an ultracold gas of 86-Sr atoms. Strong ground-state atom-atom interactions in this system influence the molecular states that are accessible and facilitate the observation of rotational structure. The large size of ULRRMs leads to a breakdown of the Franck-Condon principle. Results are interpreted with the aid of theory that accounts for the large s-wave scattering length and recoil momentum associated with photoexcitation. Several puzzles still remain unexplained, such as the sensitivity of the photoexcitation spectrum to magnetic field.

## How many ground state atoms can a Rydberg atom accommodate?

**Jan-Michael Rost**

Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

From [1] we know that a Rydberg electron can form a molecular type of orbital with a ground state atom nearby. Extended to include several atoms polyatomic Rydberg molecules in many facets [2], including scarring of Rydberg electron densities by classical periodic orbits in a dense (random) gas of atoms [3], we ask here what happens if we increase the number of (ground state) atoms in the volume of the Rydberg atom to infinity. We do this in two ways, (i) by keeping the Rydberg excitation fixed increasing the atom density [4], and (ii) by increasing the Rydberg excitation in parallel with the number of atoms producing a thermodynamic limit for a single (Rydberg) atom. The results will be presented and their implications for the quantum-classical correspondence discussed.

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## The life and fate of ultra-long-range Rydberg molecules

**Herwig Ott**

Department of Physics and Research Center OPTIMAS, Technische Universität Kaiserslautern,  
Germany

The creation of ultra-long-range Rydberg molecules in an ultracold dense atomic gas combines aspects of physical chemistry, few- and many-body physics. We use this experimental platform to create different types of Rydberg molecules with striking physical properties. We discuss the photo-association of those molecules, their interaction with the surrounding ground state atoms, their use for engineering many-body physics and their decay processes. For the latter, we have built a dedicated reaction microscope, which allows us to measure the momentum distribution of the decay products, thus revealing the internal molecular dynamics.

## Exploring the binding of long-range Rydberg molecules

**Michael Peper and Johannes Deiglmayr**

Department of Physics, University of Leipzig, DE-04109 Leipzig, Germany

In this talk, I will present an experimental toolbox for selectively varying the contributions to the binding in long-range Rydberg molecules (LRMs). By studying photoassociation of heteronuclear LRMs in a dual-species MOT of  $^{39}\text{K}$  and  $^{133}\text{Cs}$  atoms, we explore the role of the p-wave scattering and hyperfine interaction to the binding energy and stability of LRMs. Millimeter-wave spectroscopy allows us to probe the bond length and electronic character. This work paves the way to a rigorous test of theoretical models, a prerequisite for the extraction of accurate low-energy electron scattering phase shifts from experimentally observed binding energies.



## Polyatomic ultralong range Rydberg molecules

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In cold and ultracold mixtures of atoms and molecules, Rydberg interactions with surrounding atoms or molecules may, under certain conditions, lead to the formation of special long-range Rydberg molecules [1-3]. These exotic molecules provide an excellent toolkit for manipulation and control of interatomic and atom-molecule interactions, with applications in ultracold chemistry, quantum information processing and many-body quantum physics.

In this talk, we will first discuss ultralong-range polyatomic Rydberg molecules formed when a heteronuclear diatomic molecule is bound to a Rydberg atom [3,4]. The binding mechanism appears due to anisotropic scattering of the Rydberg electron from the permanent electric dipole moment of the polar molecule. We propose an experimentally realizable scheme to produce these triatomic ultralong-range Rydberg molecules in ultracold KRb traps, which might use the excitation of potassium or rubidium [5]. By exploiting the Rydberg electron-molecule anisotropic dipole interaction, we induce a near resonant coupling of the non-zero quantum defect Rydberg levels with the KRb molecule in an excited rotational level. This coupling enhances the binding of the triatomic ultralong-range Rydberg molecule and produces favorable Franck-Condon factors.

Another type of ultralong-range Rydberg molecule is formed in collisions between polar molecules in cold and ultracold settings [6]. The interaction of  $\Lambda$ -doublet nitric oxide (NO) with long-lived Rydberg NO molecules forms ultralong-range Rydberg bimolecules with GHz energies and kilo-Debye permanent electric dipole moments. The description includes both the anisotropic charge-molecular dipole interaction and the electron-NO scattering. The rotational constant for the Rydberg bimolecules is in the MHz range, allowing for microwave spectroscopy of rotational transitions in Rydberg bimolecules. The Rydberg molecules described here hold promise for studies of a special class of long-range bimolecular interactions.

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## Double Rydberg molecules

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Based on their dispersion coefficients Cote et al. suggested the existence of macrodimers, or double Rydberg molecules. Later Kiffner et al. suggested that bound molecules could be formed between two asymptotic states. The first experimental evidence for bound double Rydberg molecules was presented by Overstreet et al. Saffmanhausen and Deiglmayr, and Hollerith et al provided spectroscopic evidence for these molecules. Here we discuss microwave resonance transitions between pairs of Rydberg atoms, double Rydberg molecules, although they are not necessarily bound. Rb atoms in a magneto optical trap are excited to a Rydberg state with a narrow band pulsed laser, after which a microwave field drives the molecular transition, for example the transition  $ndnd + hv \rightarrow (n + 1)d(n - 2)f$ . This transition is most easily understood as being due to the admixture of the  $(n + 2)p(n - 2)f$  state into the  $ndnd$  state by the dipole-dipole interaction, which allows the single photon microwave transition to the final  $(n + 1)d(n - 2)f$  state to be driven. Alternatively, the transition can be treated as a Forster resonance of dressed or Floquet states. While the basic notions of these transitions are understood, there remain several questions. One is the width of the microwave resonance. What determines it? The process  $nsns + 2hv \rightarrow np_{3/2}np_{1/2}$  provides a clue. It is broadened asymmetrically to lower frequency, presumably due to the van der Waals interaction with the energetically nearby  $ns(n + 1)s$  state. At the average separation of the trap this interaction is negligible, but for the pair of closely spaced atoms undergoing the microwave transition, it is not. In the  $ndnd \rightarrow (n + 1)d(n - 2)f$  case there is a previously ignored resonant final state dipole-dipole interaction, with the  $(n - 2)f(n + 1)d$  state, which leads to the observed broadening. The  $nsns + 2hv \rightarrow np_{3/2}np_{1/2}$  case presents as well a fascinating question. In theory, the transition is only possible due to the three dimensional nature of the dipole-dipole interaction, that is to the fact that the microwave field is not aligned with the molecular axis. In a lattice it should be possible to verify or refute this claim.

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## Spectroscopy and dynamics of an ion within a Rydberg-electron orbit

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Atoms and molecules excited to a Rydberg state possess one electron residing predominantly far away from the ion core. For sufficiently large values of the principal and/or orbital-angular-momentum quantum numbers of the Rydberg electron, the ion core can be considered as isolated from the Rydberg electron. Its structure and dynamics are, to a good approximation, the same as the bare ion. This property lies, for example, at the heart of the isolated core excitation (ICE) technique widely used to study the core-excited Rydberg states of alkaline-earth-metal atoms.

In this talk, I will discuss how electronically excited states of molecular ions can be studied by probing the isolated ion core of a molecule in a high Rydberg state. High-resolution spectroscopy techniques have been developed for this purpose and include action spectroscopy schemes based on molecular photodissociation or quantum-control multiphoton schemes. These techniques were used to carry out the first characterization of the Rydberg states of a molecular cation, with the example of the  $\text{MgAr}^+$  ion. The Rydberg states of molecular ions are poorly known compared to the Rydberg states of neutral molecules, despite the fact that the former play an important role for high-resolution photoelectron spectroscopy of cations and act as transient resonances in ion-neutral collisions. Our study paves the way to systematic investigations of the Rydberg states of molecular cations.

When the ion core within the Rydberg-electron orbit is excited to a sufficiently high electronic state, it can no longer be considered as isolated from the Rydberg electron and electronic correlations play an important role. As noted by others, the transition from the isolated-core regime to the regime where correlations are important offers interesting insights into electronic correlations. Electronic states of Mg and Sr atoms in which both of their valence electrons are highly excited are ideal to study these correlations. I will present the results of joint experimental and theoretical investigations of the so-called planetary states of Mg and Sr, with particular emphasis on the role played by correlations in the two-electron dynamics.

# Quantum defects for molecular Rydberg states and precision spectroscopy of $\text{H}_2^+$

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Precision spectroscopy of high molecular Rydberg states represents a powerful method for the determination of ionization energies and of rovibronic intervals of molecular ions. In this context, the one- and two-electron molecules  $\text{H}_2^+$  and  $\text{H}_2$  are particularly attractive because their energy levels can be calculated with extraordinary accuracy, allowing to test molecular quantum electrodynamics (QED), search for new physics beyond the Standard Model and determine fundamental constants.

The bound energy levels of the Rydberg states are obtained by multichannel quantum-defect theory (MQDT), with the information of the Rydberg electron-ion collision being contained in the quantum defects, which depend on the quantum state of the ion.

It will be shown, that using very accurate Born-Oppenheimer potential energy curves for low- $nf$  Rydberg states ( $n = 4 - 7$ ), ab initio quantum-defect functions dependent on the internuclear distance, the projection of the orbital angular momentum  $\Lambda$ , the total electron spin and the energy of the Rydberg electron can be obtained.

Using the concept of a frame transformation, these body-fixed quantum defects allow to describe the collision of the Rydberg electron with the molecular ion core including spin-rovibrational degrees-of-freedom, as well as relativistic and QED effects with an accuracy much better than a MHz.

As an example, it will be shown how the new quantum defects allow the determination of the fundamental vibrational interval of  $\text{H}_2^+$  with unprecedented accuracy, using experimental data of non-penetrating  $f$  Rydberg states.

## Vibrational autoionization of core-nonpenetrating Rydberg states of NO: Long-range *ab initio* model and applications to cold ion chemistry

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High orbital angular momentum ( $\ell \geq 3$ ), core-nonpenetrating Rydberg states can be well described by a simplified model in which the Rydberg electron is only weakly perturbed by the long-range electric properties (i.e., multipole moments and polarizabilities) of the ion-core. We have used a long-range model to describe the vibrational autoionization dynamics of high- $\ell$  Rydberg states of nitric oxide (NO). Our model explains the extensive angular momentum exchange between the ion-core and Rydberg electron that had been previously observed in vibrational autoionization of  $f(\ell = 3)$  Rydberg states. These results shed light on a long-standing mechanistic question around these previous observations, and support a direct, vibrational mechanism of autoionization over an indirect, predissociation-mediated mechanism. In addition, our model correctly predicts newly measured total decay rates of  $g(\ell = 4)$  Rydberg states because, for  $\ell \geq 4$ , the non-radiative decay is dominated by autoionization rather than predissociation. We examine the predicted  $\text{NO}^+$  ion rotational state distributions generated by vibrational autoionization of  $g$  states and find qualitatively different behavior for these higher  $\ell$  states. Finally, we discuss generalizations of our model to other molecular systems and applications to achieve quantum state selection in the production of molecular ions.

## Effects of weak rotational and vibrational interactions on the lifetimes of long-lived Rydberg states of NO

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High Rydberg states of small molecules play an important role in electron-ion recombination in laboratory, atmospheric, and astrophysical plasmas [1-3]. However, until recently laboratory studies of the slow decay dynamics of molecules in these excited states have not been possible [4,5]. Here we describe a unique, state-of-the-art cryogenically cooled chip-based Rydberg-Stark decelerator, and the use of this device to electrostatically trap nitric oxide (NO) molecules for the first time [6]. Trapping the molecules, prepared in long-lived Rydberg states using resonance-enhanced two-colour two-photon excitation from the  $X^2\Pi_{1/2}$  ground state, for up to 1 ms enabled precise measurements of excited state decay rates [6,7]. These measurements were performed for molecules photoexcited to Rydberg states with principal quantum numbers,  $n$ , between 32 and 50, in Rydberg series converging to the  $N^+ = 0, 1$ , and 2 rotational states of the  $v^+ = 0$  vibrational state of  $\text{NO}^+$ . For the range of Rydberg states studied, decay time constants between 200  $\mu\text{s}$  and 400  $\mu\text{s}$  were observed to generally decrease as the value of  $n$  was increased. However, for some particular values of  $n$  deviations from this trend were seen. Therefore the long-lived states studied in these experiments did not follow the typical  $n$ -scaling rules of high Rydberg states. With the aid of numerical calculations of the energy-level structure and lifetimes of the Rydberg states, these observations were interpreted to arise as a result of weak rotational and vibrational channel interactions. The vibrational channel interactions, between Rydberg states in the  $v^+ = 0$  vibrational series and nearby short-lived ( $\sim 1$  ps) low- $n$  ( $n = 7$ ) states in the  $v^+ = 1$  series, resulted in contributions to the total excited-state decay rates of  $\sim 1$  kHz. Rotational channel interactions, within the  $v^+ = 0$  series, were identified to specifically affect the decay time constants of some individual Rydberg states.

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**Part II**

**Posters**

# Self-assembling of the intermediate with chemically bound argon in van der Waals complex Ar-I<sub>2</sub> photoexcited into the Rydberg states of I<sub>2</sub>

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The attention to the chemical compounds of noble gases Ng is dictated by the fundamental interest in the nature of the bonding of these inert atoms. Moreover, the bonding motifs which make the neutral compounds of noble gases stable can provide stability of the variety of the new currently unexpected substances of other elements as well. The chemical inertness increases for lighter Ng atoms. Up to now only one neutral compound containing chemically bound argon has been synthesized. This is hydride HArF [1]. The results obtained in the presented work reveal photogeneration of the other type of Ar compound. In our experiments the photodissociation of van der Waals complex Ar-I<sub>2</sub> provided by the excitation of I<sub>2</sub> into Rydberg states has been investigated. Velocity map imaging of the photofragments allowed us to reveal the formation of translationally hot Ar<sup>+</sup> ions. Three modes in the kinetic energy distribution of these ions indicate three photodissociation channels giving rise to Ar<sup>+</sup>. The measured kinetic energy of ions and its dependence on the energy of pumping photons allow us to conclude formation of neutral linear intermediate Ar<sup>+</sup>-I-I<sup>-</sup> being a source of Ar<sup>+</sup> ions. This neutral compound is isoelectronic to trihalide anion Cl-I-I<sup>-</sup>. The energetic characteristics of appearing Ar(+) ions correspond very well to the photodissociation channels expected for Ar<sup>+</sup>-I-I<sup>-</sup> by analogy with trihalide anions. The self-assembling of the configuration making the formation of this compound possible is confirmed by modeling the dynamics in the excited linear complex Ar-I<sub>2</sub>. The excitation state giving rise to this compound is the ion-pair state of I<sub>2</sub> which is known [2] to follow Rydberg excitation of free iodine in this spectral region. The obtained results allow us to suppose that the excitation of the van der Waals complexes of noble gases with halogens into the Rydberg states of halogen is a promising approach for generating neutral compounds containing chemically bound atoms of noble gases. The results to be presented are published recently [3].

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# Ionization energy of the metastable $2^1S_0$ state of $^4\text{He}$ from Rydberg-series extrapolation

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In a recent breakthrough in first-principles calculations of two-electron systems, Patkóš, Yerokhin and Pachucki [PRA **103**, 042809 (2021)] have performed the first complete calculation of the Lamb shift of the helium  $2^3S_1$  and  $2^3P_J$  triplet states up to the term in  $\alpha^7 m$ . Whereas their theoretical result of the frequency of the  $2^3P \leftarrow 2^3S$  transition perfectly agrees with the experimental value, a more than  $10\sigma$  discrepancy was identified for the  $3^3D \leftarrow 2^3S$  and  $3^3D \leftarrow 2^3P$  transitions, which hinders the determination of the  $\text{He}^{2+}$  charge radius from atomic spectroscopy that is necessary to complement the recent  $\alpha$ -particle charge radius determination using muonic helium from J. Krauth et al. [Nature **589**, 527531 (2021)].

We report on the determination of the ionization energy of the metastable  $2^1S_0$  state of He (960332040.491(32) MHz) by Rydberg-series extrapolation through the determination of the frequencies of 21 transitions from the  $2^1S_0$  state to  $np$  Rydberg states with principal quantum number  $n$  in the range between 24 and 102, yielding a relative uncertainty of  $3 \times 10^{-11}$  [PRL **127**, 093001 (2021)]. A one-photon ( $\sim 312$  nm) excitation scheme is used for Rydberg-state excitation of metastable He atoms in a doubly skimmed supersonic beam. The absolute frequency calibration is achieved using a frequency comb referenced to a GPS-disciplined Rb clock.

This absolute measurement is used in combination with the  $2^3S_1 \leftarrow 2^1S_0$  interval measured by van Rengelink et al. [Nat. Phys. **14**, 1132 (2018)] and the  $2^3P \leftarrow 2^3S_1$  interval measured by Zheng et al. [PRL **119**, 263002 (2017)] and Cancio Pastor et al. [PRL **92**, 023001 (2004)] to derive experimental ionization energies of the  $2^3S_1$  state (1152842742.640(32) MHz) and the  $2^3P$  centroid energy (876106247.025(39) MHz). These values reveal disagreements with the  $\alpha^7 m$  Lamb shift prediction by  $6.5\sigma$  and  $10\sigma$ , respectively, and support the suggestion by Patkóš et al. of an unknown theoretical contribution to the Lamb shifts of the  $2^3S$  and  $2^3P$  states of He.

# Dissipative dynamics of a molecular Rydberg gas: Avalanche to an ultracold plasma state of strong coupling

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We predict the existence of a universal class of ultralong-range Rydberg molecular states whose vibrational spectra form trimmed Rydberg series. A dressed ion-pair model captures the physical origin of these exotic molecules, accurately predicts their properties, and reveals features of ultralong-range Rydberg molecules and heavy Rydberg states with a surprisingly small Rydberg constant. The latter is determined by the small effective charge of the dressed anion, which outweighs the contribution of the molecule's large reduced mass. This renders these molecules the only known few-body systems to have a Rydberg constant smaller than  $R_\infty/2$ .

# Towards a hybrid platform of ultracold polar molecules and Rydberg atoms

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The rich internal structure and intrinsic dipolar interactions possessed by polar molecules makes them a promising system for exploring quantum chemistry, quantum computation and quantum simulation. However, to fully realise their potential in these areas, single-site control and detection of molecules is desirable. Such control is natively realised using optical tweezer arrays to confine the molecules, which enables trapping and rearrangement of the particles to create arrays in flexible geometries.

In our experiment [1], we aim to produce single RbCs molecules in optical tweezers. The molecules will be formed by magnetoassociation of Rb and Cs atoms prepared in the motional ground state of the tweezer traps [2]. I will present recent results demonstrating cooling of Rb and Cs atoms using Raman sideband cooling, where we obtain 3D ground state fractions of 0.86(5) and 0.94(2) respectively. By utilising excess Rb atoms prepared in the motional ground state and exciting them to Rydberg states we aim to realise a hybrid system which combines arrays of ultracold molecules and Rydberg atoms. I will discuss our progress towards this goal and describe how we plan to use this hybrid system to photoassociate giant polyatomic Rydberg molecules [3,4] and test schemes for quantum non-demolition readout of the diatomic molecule's quantum state [5].

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# Low-energy reactive collisions of H<sub>2</sub>, HD, and D<sub>2</sub> with H<sub>2</sub><sup>+</sup>, HD<sup>+</sup>, D<sub>2</sub><sup>+</sup>: branching ratios, deviation from Langevin behavior and kinetic energy analysis of the products.

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The study of low-temperature collisions between small molecules, molecular hydrogen in particular, is crucial to understand the chemistry of interstellar clouds, characterized by temperatures typically in the 3 K to 60 K range. These studies can also be used to test theoretical predictions about the reaction rates, the product branching ratios, and the product kinetic-energy distributions [1]. The ion-molecule capture rates are well described by the semi-classical Langevin theory at high to moderate energy, but deviations at low energies are predicted, and experimentally observed [1,2,3].

Low-energy collisions of molecules with ions are difficult to study because stray fields heat the ions up. This difficulty is circumvented by replacing the ions with the corresponding parent neutral molecule in highly excited Rydberg states. The Rydberg electron, very far from the core and loosely bound, shields the ion core from external fields while having negligible impact on its reactions with neutral molecules located within the Rydberg-electron orbit [3,4]. To reach low collision energies, velocity-tunable supersonic beams of the reactants are merged.

Rydberg-Stark states are produced in one of the two beams by photoexcitation in the presence of an electric field and subsequently velocity selected and deflected using a curved chip-based surface-electrode deflector [5]. This deflector exploits the very large dipole moments of Rydberg-Stark states and allows us to merge the Rydberg molecular beam with a supersonic beam of the ground-state neutral molecules. The relative and absolute axial velocities of the reactants can also be varied by adjusting the velocity selected by the deflector, by varying the delays between the openings of the valves, and by changing their temperatures. The longitudinal velocity spread of the molecular beams rapidly evolve into a spatial dispersion, which enables a high collision-energy resolution and studies at collision energies below  $k_B \cdot 1$  K, where the reaction rates are enhanced relative to the Langevin capture rate for a pure ion-induced-

dipole interaction [1,2].

We report on experimental studies of low-energy ion-molecule reactions between the neutral molecules  $\text{H}_2$ ,  $\text{HD}$ , and  $\text{D}_2$  and the molecular ions  $\text{H}_2^+$ ,  $\text{HD}^+$ , and  $\text{D}_2^+$ . Differences resulting from nuclear-spin symmetry were investigated by exchanging natural  $\text{H}_2$  with para-enriched  $\text{H}_2$ . These studies allow systematic quantitative analysis of the enhancement effect for systems of different reduced masses and rotational-state occupations of the ground-state reactant. The observations can be explained by ion-quadrupole interaction and quantum-capture contributions. The studies also allow the determination of the branching ratios between different product channels, i.e.,  $\text{H}_2\text{D}^+ + \text{H}$  and  $\text{H}_3^+ + \text{D}$  in the reaction of  $\text{H}_2$  with  $\text{HD}^+$ , to test simple theoretical models, such as a combinatorial model, a model describing the reactions as H-,  $\text{H}^+$ -, D-, and  $\text{D}^+$ -transfer processes, and a statistical model relating the reaction rate coefficients to the translational and rovibrational state densities of the product channels.

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# Spectroscopic studies of H<sub>2</sub>S and D<sub>2</sub>S near ion-pair dissociation thresholds

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We present the results of spectroscopic investigations of ion-pair states of small polyatomic molecules near ion-pair thresholds. These states are bound states of an attractive Coulomb potential between the positively and negatively charged fragments. They are analogous to high Rydberg states of atoms and molecules, but with a much larger reduced mass. They are therefore called heavy Rydberg states. Such heavy Rydberg states are well characterized in diatomic molecules [1,2] but have not been observed yet in many polyatomic molecules [3,4] so that many of their characteristics are not well understood. Accurate threshold energies give access to thermochemical properties such as bond energies. We are currently investigating triatomic systems, in particular H<sub>2</sub>S and D<sub>2</sub>S. The sample of interest is cooled down in a supersonic expansion. The cold gas cloud passes a skimmer before it is excited via a one-photon transition with narrow-band VUV laser radiation to high-lying heavy Rydberg states with principal quantum numbers  $n \simeq 10000$ . After delayed pulsed-field dissociation, the molecular fragments are extracted in a photoion/photoelectron time-of-flight spectrometer. We determined the ion-pair formation energy for the two lowest thresholds of the SH<sup>-</sup> X<sup>1</sup>Σ<sup>+</sup>( $v^- = 0, 1$ ) + H<sup>+</sup> ion-pair channel to be  $E_{\text{IP}}^{\text{H}_2\text{S}}(v^- = 0, N^- = 0) = 122460 \pm 2 \text{ cm}^{-1}$  and  $E_{\text{IP}}^{\text{H}_2\text{S}}(v^- = 1, N^- = 0) = 124997 \pm 5 \text{ cm}^{-1}$ , respectively. For the SD<sup>-</sup> X<sup>1</sup>Σ<sup>+</sup>( $v^- = 0$ ) + D<sup>+</sup> channel we found  $E_{\text{IP}}^{\text{D}_2\text{S}}(v^- = 0, N^- = 0) = 123035 \pm 5 \text{ cm}^{-1}$ . With this values, we can improve the bond energy [ $D_0(\text{HS} - \text{H})$  and  $D_0(\text{DS} - \text{D})$ ] of the neutral species of both molecules.

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# Self-organization in the avalanche, quench and dissipation of a molecular ultracold plasma

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Spontaneous avalanche to plasma splits the core of an ellipsoidal Rydberg gas of nitric oxide. Ambipolar expansion first quenches the electron temperature of this core plasma. Then, long-range, resonant charge transfer from ballistic ions to frozen Rydberg molecules in the wings of the ellipsoid quenches the centre-of-mass ion/Rydberg molecule velocity distribution. This sequence of steps gives rise to a remarkable mechanics of self-assembly, in which the kinetic energy of initially formed hot electrons and ions drives an observed separation of plasma volumes. These dynamics adiabatically sequester energy in a reservoir of mass transport, starting a process that anneals separating volumes to form an apparent glass of strongly coupled ions and electrons. Short-time electron spectroscopy provides experimental evidence for complete ionization. The long lifetime of this system, particularly its stability with respect to recombination and neutral dissociation, suggests that this transformation affords a robust state of arrested relaxation, far from thermal equilibrium. We see this most directly in the absorption spectrum of transitions to states in the initially selected Rydberg series, detected as the long-lived signal that survives a flight time of 400  $\mu\text{s}$  to reach an imaging detector. The initial density of electrons produced by prompt Penning ionization, which varies with the selected initial principal quantum number and density of the Rydberg gas, determines a balance between the rising density of ions and the falling density of Rydberg molecules. This Penning-regulated ion–Rydberg-molecule balance appears necessary as a critical factor in achieving the long ultracold plasma lifetime to produce spectral features detected after very long delays.

# Test of the spectator role of the Rydberg electron in the $\text{He}(n) + \text{CO}$ reaction and effects of the CO dipole and quadrupole moments at low collision energies

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Rydberg atoms have large electric dipole moments and therefore their translational motion can be easily manipulated by inhomogeneous electric fields [1-3]. We deflect a beam of He Rydberg atoms using a Rydberg-Stark surface deflector and accelerator and merge it with a beam of ground-state CO molecules to study the  $\text{He}^+ + \text{CO} \rightarrow \text{C}^+ + \text{O} + \text{He}$  reaction. In the experiment, we exploit the facts that the Rydberg electron prevents the heating up of the ions by stray electric fields and that it hardly affects the ion-molecule reaction taking place within its orbit [4-8], in accordance with the independent particle model of Rydberg collisions [9,10]. We detect the  $\text{C}^+$  product of the reaction as a function of the velocity of the  $\text{He}(n)$  atoms. In this way, we can adjust the collision energy of the reaction and probe the range between 0 and  $\sim k_B \cdot 25$  K.

In the first part of our investigation, we tested the spectator role of the Rydberg electron by measuring the distribution of principal quantum numbers of the  $\text{He}(n)$  reactants and the  $\text{C}(n')$  products using pulsed-field ionization [11]. We find that the distributions are affected by spontaneous emission and blackbody-radiation-induced transitions, but not by the ion-molecule reaction, which in turn indicates that the Rydberg electron does not influence the ion-molecule reaction. In the second part, we carried out an investigation of the effect of the dipole and quadrupole moments of CO on the reaction rates at low collision energies. In particular, we observe a 30% decrease in the product formation at the lowest collision energies (below  $\sim k_B \cdot 5$  K) and attribute it to the negative quadrupole moment of CO on the basis of calculated state-dependent capture-rate coefficients.

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# Non-adiabatic dynamics in Rydberg gases with random atom positions

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Assemblies of highly excited Rydberg atoms in an ultracold gas can be set into motion by a combination of van-der-Waals and resonant dipole-dipole interactions [1]. Thereby, the collective electronic Rydberg state might change due to non-adiabatic transitions, in particular if the configuration encounters a conical interaction [2]. For the experimentally most accessible scenario, in which the Rydberg atoms are initially randomly excited in a three-dimensional bulk gas under blockade conditions, we numerically show that non-adiabatic transitions can be common when starting from the most energetic repulsive BO-surface [3]. We outline how this state can be selectively excited using a microwave resonance, and demonstrate a regime where almost all collisional ionization of Rydberg atoms can be traced back to a prior non-adiabatic transition. Since Rydberg ionisation is relatively straightforward to detect, the excitation and measurement scheme considered here renders non-adiabatic effects in Rydberg motion easier to demonstrate experimentally than in scenarios considered previously [4,5].

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# Multipolar character of Förster resonance energy transfer

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Förster resonance energy transfer (FRET) refers to non-radiative exchange of energy between two particles, typically described in terms of resonant dipole-dipole interaction. When the separation between the particles becomes comparable to their size, however, the dipole approximation breaks down. This is expected for Rydberg atoms which possess large dipole moments and are thus ideal for the study of FRET, for example, when colliding them with polar molecules [1,2].

Here, we study the multipolar character of FRET between Rydberg atoms and polar molecules. To this end, we develop a theoretical model for describing the scattering process considering dipole-dipole, dipole-quadrupole and dipole-octupole interactions. We determine the breakdown of the dipole approximation and assess the role of higher order contributions by computing the scattering cross sections. Moreover, we suggest to control the cross section in experiments by making use of the Stark effect of the Rydberg atom.

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# Imaging the interface of a qubit and its quantum-many-body environment

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We show that two major facets of the decoherence paradigm are experimentally accessible for a single impurity atom embedded in a Bose-Einstein condensate when the impurity is brought into an electronic superposition of two Rydberg states [1]. Not only can the electronic decoherence of the Rydberg atom be read out by microwave interferometry [2,3], the platform also provides unique access to the accompanying entangled state of the environment. We theoretically demonstrate signatures of the latter in total atom densities during the transient time in which the impurity is becoming entangled with the medium but the resultant decoherence is not complete yet. The Rydberg impurity thus provides a handle to initiate and read-out mesoscopically entangled superposition states of Bose atom clouds affecting about 500 condensate atoms. We find that the time-scale for its creation and decoherence can be tuned from the order of nanoseconds to microseconds by choice of the excited Rydberg principal quantum number  $\nu$  and that Rydberg decoherence dynamics is typically non-Markovian [4].

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# Control of molecular ultracold plasma relaxation dynamics in mm-wave and radio-frequency fields

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Resonant mm-wave fields in the range from 50 to 100 GHz drive  $f \rightarrow g$  transitions in a state-selected  $nf(2)$  Rydberg gas of NO. This transformation produces a clear signature in the selected field ionization (SFI) spectrum and dramatically increases the early time intensity of high-Rydberg resonances in the SFI-detected high-Rydberg spectrum. We associate these enhanced features with a decrease in the rate of predissociation owing to an increase in Rydberg orbital angular momentum. This same signature of early time Rydberg stabilization appears as an enhanced long-time plasma signal, extracted as a late peak after 20  $\mu$ s of field-free evolution. We can conclude from these observations that avalanche alone does not guarantee a plasma state of arrested relaxation. But rather, the formation of an arrested phase requires both avalanche-produced nitric oxide ions and a persistent population of long-lived Rydberg molecules. A 250 ns 60 MHz radio frequency pulse with a peak-to-peak amplitude as low as 400 mV/cm, applied with zero delay similarly increases the signal of associated with a residue of lower- $n$  Rydberg molecules detected microseconds later. Applied later to a plasma in a state of arrested relaxation, however, the same radiofrequency field depletes the residual Rydberg signal. We associate both effects with Rydberg electronic orbital angular momentum mixing. At early times the applied field mixes the photoselected  $nf$  state mixes with longer-lived states of high angular momentum. Later, electrons released by the radio frequency field collide with Rydberg molecules trapped in states of high angular momentum driving a predissociative flux through channels of low  $\ell$ .



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