

SUMMER SCHOOL 2019 IRTG CoCo Cold Controlled Ensembles in Physics and Chemistry



BOOK OF ABSTRACTS

4th CoCo Summer School

Cold Molecules and their Interactions

Theoretical and Experimental Methods

July 29 – August 03, 2019

Okanagan Campus, Kelowna, Canada





Impressum

IRTG 2079 / Cold Controlled Ensembles in Physics and Chemistry Institute of Physics University of Freiburg Hermann-Herder-Straße 3 79104 Freiburg http://www.irtg-coco.uni-freiburg.de



Main Topics

Sergey Bokarev

Electronic structure theory for photophysics of isolated molecules

Michael Tarbutt

Formation and application of cold and ultracold molecules

Gary Douberly

Aggregation in Helium Droplets

Olivier Dulieu

Theory of cold molecules

James P. Shaffer

Rydberg Molecules, Interactions and Applications

Financial Support





Organisation

IRTG 7079 "Cold Controlled Ensembles in Physics and Chemistry"

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General Information

Duration

Monday, July 29, 201, 4:00 p.m. – Saturday, August 3, 2019, 5:00 p.m.

Location

UBC Okanagan Campus Engineering, Management and Education (EME) - 2141 1137 Alumni Avenue, Kelowna, BC V1V 1V7, Canada

Accommodation

Conferences & Accommodation, Okanagan Campus

1290 International Mews

Kelowna, BC Canada V1V 1V8

www.okanagan.ubcconferences.com

<u>Adjoining single rooms for doctoral students</u>: 2 bedrooms with a single bed in each air-conditioned room and a private washroom. Each floor has a common lounge with kitchenette and comfortable seating and TV.

<u>Studio Suites for speakers and PIs</u>: provides 1 queen bed, a private washroom and a basic equipped kitchen with a small dining area.

 \rightarrow The rooms will be set up with towels and linens.

Travel Information - Shuttles

BY AIR & TAXI

UBC's Okanagan campus is across from the Kelowna International Airport. Please take a taxi (around \$12 one way) to our 24/7 Front Desk on 1290 International Mews (Nicola Residence).



BY PUBLIC TRANSPORT

Various buses stop on campus. When arriving on campus (at the transit loop), walk north on Alumni Drive, then left onto University Way, then take a right at International Mews. The Front Desk is located in the second residence building on your left, called Nicola Residence.





Meals & Drinks

Meals and drinks during the scientific program are included in the conference fee. We will inform you on the spot where the meals will take place.

Excursion

On Wednesday, July 31, an excursion is organized. We will start after a short lunch break and drive by bus to Kalamalka Lake Provincial Park for a hiking tour. Afterwards we visit the Mission Hill Estate Winery. There will be a guided tour, wine tasting and dinner.



(Kalamalka Lake Provinial Park)



Free time activities

Saturday evening, after the Closing Session of the Summer School, is free time. No dinner or event is planned.

WiFi

There is WiFi access throughout accommodation area.

Travel to Vancouver on Sunday, August 4

After breakfast, there will be a bus shuttle to Vancouver. On the way, we will make an excursion and hike through the Othello tunnels. A lunch box will be provided for each participant.



(Othello Tunnels, Hope)



Program of the IRTG Summer School 2019

Monday, July 29, 2019

4:00 p.m.	Registration			
4:30 p.m.	Takamasa Momose & Michael Walter			
	Welcome and Introduction			
7:30 p.m.	Dinner			

Tuesday, July 30, 2019

9:00 a.m.	Sergey Bokarev Electronic structure theory for photophysics of isolated molecules				
10:30 a.m.	Break				
11:00 a.m.	Sergey Bokarev				
	Electronic structure theory for photophysics of isolated molecules				
12:30 p.m.	Lunch Break				
2:30 p.m.	Sergey Bokarev				
	Electronic structure theory for photophysics of isolated molecules				
4:00 p.m.	Break				
4:30 p.m.	Michael Tarbutt				
	Formation and application of cold and ultracold molecules				
7:30 p.m.	Dinner				

Wednesday, July 31, 2019

9:00 a.m.	Michael Tarbutt			
	Formation and application of cold and ultracold molecules			
10:30 a.m.	Break			
11:00 a.m.	Michael Tarbutt			
	Formation and application of cold and ultracold molecules			
12:30 p.m.	Lunch Break			
2:00 p.m.	Excursion: Hiking Tour, Winery visit & Dinner			



Program of the IRTG Summer School 2019

Thursday, August 1, 2019

9:00 a.m.	Gary Douberly			
	Aggregation in Helium Droplets			
10:30 a.m.	Break			
11:00 a.m.	Gary Douberly			
	Aggregation in Helium Droplets			
12:30 p.m.	Lunch Break			
2:30 p.m.	Gary Douberly			
	Aggregation in Helium Droplets			
4:00 p.m.	Break			
4:30 p.m.	Scientific Karaoke, Session I (PhD internal)			
7:30 p.m.	Dinner			

Friday, August 2, 2019

9:00 a.m.	Olivier Dulieu					
	Theory of ultracold molecules					
10:30 a.m.	Break					
11:00 a.m.	Olivier Dulieu					
	Theory of ultracold molecules					
12:30 p.m.	Lunch Break					
2:30 p.m.	Olivier Dulieu					
	Theory of ultracold molecules					
4:00 p.m.	Break					
4:30 p.m.	Scientific Karaoke, Session II (PhD internal)					
7:30 p.m.	Dinner					



Program of the IRTG Summer School 2019

Saturday, August 3, 2019

9:00 a.m.	James P. Shaffer					
	Rydberg Molecules, Interactions and Applications					
10:30 a.m.	Break					
11:00 a.m.	James P. Shaffer					
	Rydberg Molecules, Interactions and Applications					
12:30 p.m.	Lunch Break					
2:30 p.m.	James P. Shaffer					
	Rydberg Molecules, Interactions and Applications					
4:00 p.m.	Break					
4:30 p.m.	Closing Session					
	Takamasa Momose, Michael Walter & Giuseppe Sansone					



Abstracts – Invited Speakers



Electronic structure theory for photophysics of isolated molecules

Sergey I. Bokarev

Institute for Physics, University of Rostock, Germany

Interpretation of steady-state and time-resolved spectroscopic experiments strongly relies on the underlying physical models. Here, theoretical simulations play an increasing role in the development of these models, becoming more and more popular with the growth of the available computational power. In particular, understanding the photophysics and photochemistry of molecular species requires knowledge of potential energy surfaces of the excited electronic states. During decades, density functional theory (DFT) and its linear-response time-dependent counterpart (TDDFT) have proven themselves to be very efficient for predicting properties of condensed phases and molecules [1].

Although DFT is in principle an exact theory, it heavily relies on the exchangecorrelation energy functional. Its exact form is unknown and is usually fitted to experimental data or obtained on the basis of model systems with known properties. The approximate nature of the density functional is the origin of all the caveats and the sometimes spurious behavior of the results obtained with DFT. Two main erroneous effects introduced by approximate functionals are self-interaction and violation of the derivative discontinuity condition. These problems manifest themselves in wrong predictions of various molecular properties [2,3].

One of the examples is a severe error in energies of charge-transfer electronic states, which are very important for photophysics and photochemistry. Apart from determining the shape of low-energy absorption spectra, these states play an important role throughout different fields of science. For instance, initial charge separation constitutes the basis of natural



and artificial light-harvesting systems as applied in photovoltaics and photocatalysis.

Whenever we speak about charge-transfer electronic states, the essential parameter of the density functional, influencing the reliability of the charge-transfer excitation energies, is the content of the exact orbital-dependent exchange energy. At this point, a remedy for the deficiencies of the conventional DFT is provided by the range-separation concept introducing the exact exchange energy functional at long interelectron distances [2-4].

In this lecture, I will give a brief introduction to the electronic structure theory of isolated molecules with the focus on Kohn-Sham DFT and TDDFT theories. I will describe the fully self-consistent first principles scheme how to correct for the self-interaction and derivative discontinuity error and how it leads to reliable predictions of the properties of charge-transfer states. A series of practical tasks addressing different aspects of the range-separated DFT and its application to the excited states of model molecules and small clusters will be offered.

- 1. W. Koch and M. C. Holthausen, *A Chemist's Guide to Density Functional Theory* (Wiley-VCH, Weinheim, 2001).
- 2. S. Kümmel and L. Kronik, *Orbital-dependent density functionals: Theory and applications*, Rev. Mod. Phys. **80**, 3–60 (2008).
- **3.** R. Baer, E. Livshits, and U. Salzner, *Tuned Range-Separated Hybrids in Density Functional Theory*, Annu. Rev. Phys. Chem. **61**, 85–109 (2010).
- S. I. Bokarev, O. S. Bokareva, and O. Kühn, A theoretical perspective on charge transfer in photocatalysis. The example of Ir-based systems, Coord. Chem. Rev. 304-305, 133–145 (2015).



Formation and applications of cold and ultracold molecules

Mike Tarbutt

Centre for Cold Matter, Department of Physics, Imperial College London, GB

In the first lecture, I will explain how cold and ultracold molecules can be used to test fundamental physics, simulate many-body quantum systems, process quantum information, and study ultracold chemistry.

In the second lecture, I will explain how direct laser cooling can be applied to molecules and how these laser-cooled molecules can be captured in magnetooptical traps. In the third lecture, I will show how electric and magnetic fields can be used to decelerate and trap molecules.

Throughout, I will give an overview of the current status of this field, illustrating the principles with examples from recent or current experiments. One of your tasks will be to decide how you would like to distribute a limited budget amongst a wide range of research activities in this field.



Aggregation in Helium Droplets

Gary E. Douberly

Department of Chemistry, University of Georgia, Athens, USA

The first beam of helium droplets was reported in the 1961 paper *Strahlen aus kondensiertem Helium im Hochvakuum* by Von E. W. Becker and co-workers [1]. However, molecular spectroscopy of helium-solvated dopants wasn't realized until 30 years later in the laboratories of Scoles and Toennies [2,3].

It has now been over two decades since this early, seminal work on doped helium droplets, yet the field of helium droplet spectroscopy is still fresh with vast potential. Analogous in many ways to cryogenic matrix isolation spectroscopy, the helium droplet is an ideal environment to spectroscopically probe difficult to prepare molecular species, such as radicals, carbenes and ions. The quantum nature of helium at 0.35 K often results in molecular spectra that are sufficiently resolved to evoke an analysis of line shapes and finestructure that demands rigorous "effective Hamiltonian" treatments. The present series of lectures will focus on our successful attempts to efficiently dope organic molecular radicals and carbenes into helium droplets. These properties of these systems have been probed with infrared laser Stark and Zeeman spectroscopies. We will explore the various theoretical spectroscopic techniques applied to the analysis of such spectra, focusing on the often anomalous effects arising from the superfluid solvent environment.

- [1] E. W. Becker, R. Klingelh\"{o}fer, P. Lohse, Z. Naturforsch. A 16A, 1259 (1961).
- [2] S. Goyal, D. L. Schutt, G. Scoles, Phys. Rev. Lett. 69, 933 (1992).
- [3] M. Hartmann, R. E. Miller, J. P. Toennies, A. F. Vilesov, *Phys. Rev. Lett.* **75**, 1566 (1995).



Theory of ultracold molecules

Olivier Dulieu

Laboratoire Aimé Cotton Université Paris-Sud, Campus d'Orsay, Orsay Cedex, France

As a counterpart of Prof. M. Tarbutt's lecture, I will focus my lectures on the methods to create ultracold molecules for the association of ultracold atoms. I will first review more than 30 years of researches in this field, since the first theoretical paper published in 1987.

Then I will introduce the main concepts necessary for understanding the formation (and destruction, detection) of ultracold molecules (molecular symmetries, potential energy curves, scattering length, Feshbach resonances,...). With the help of a couple of practical cases, I will attempt to demonstrate that diatomic molecules are quantum objects, which are fun to play with.



Rydberg Molecules, Interactions and Applications

James P. Shaffer

Quantum Valley Ideas Laboratories, Waterloo, Canada

Rydberg atoms possess uniquely exaggerated properties that are useful for the development of quantum technologies. In order to fully exploit Rydberg atom characteristics, we must understand how to balance desirable properties against undesirable ones. One of the most important and celebrated properties of Rydberg atoms are their unusually long-range interactions. These interactions lead to Rydberg atom blockade phenomena and molecules with micron-sized bond lengths, but can also increase dephasing rates which are detrimental to some applications. Also unique to the system are so-called ultra long-range Rydberg molecules where the scattering of the Rydberg electron from atoms located within the Rydberg electron orbit can form a weakly bound molecule. These molecules can be dimers, trimers and higher order molecules enabling few- and many-body physics to be investigated, but can also lead to significant dephasing in some experiments. Ultra long-range diatomic Rydberg molecules, the simplest of these molecules, have exotic properties such as huge dipole moments despite being homonuclear diatomic molecules. In this series of lectures, I will introduce some basic concepts in Rydberg atom physics followed by a description of Rydberg atom interactions and the unique properties of ultra long-range Rydberg molecules. Applications will be highlighted where the properties of long-range Rydberg atom interactions are important whether they are an asset or liability. Some of most exotic properties of Rydberg molecules will be explained and illustrated. We will address both experimental and theoretical perspectives.



Abstracts – Scientific Karaoke



Photoisomerization of azobenzene and its substituted derivatives:

the effect of Protonation

Meral Ari

Institute of Physics, University of Freiburg, Germany

One of the most used organic chromophores for optical switching applications are azobenzenes. Azobenzenes exhibit a reversible isomerisation process between its trans and cis isomers of different stability. After a photochemical conversion, the spontaneous thermal back reaction occurs. While the photoinduced trans-to-cis isomerisation reaction can be performed in a few femtoseconds, the rate of the thermal cis-to-trans back reaction depends greatly on the chemical architecture of the system and the environment [1]. While slow thermally back-isomerising azoderivatives are valuable photoactive basic materials for information storage purposes, transmitting at the molecular scale with response times within the nanosecond or picosecond range will enable the potential application of azobenzene-based materials in micropumps and autonomous valves. Novel absorption spectroscopy experiments have shown that the rate of thermal back-isomerization is strongly dependent on pH [2].

With the purpose of understanding the mechanism underlying the isomerization and the changes in reaction paths and energy barriers due to protonation, in collaboration with the group of Andreas Walther from the Institute of Macromolecular Chemistry in Freiburg, we investigated the protonation sites of an azo-derivative (Arylazopyrazole structure (AAP)). Large effects on back reaction rates were observed experimentally. We computationally explored the relative energy profiles for the isomerization along the CNNC-dihedral angle of unprotonated and protonated AAP. We've



found out that protons clearly prefer to be placed at the azo-bridge and while unprotonated form prefers rotation path for the back-isomerization, the protonated form prefers inversion. The effect of different substitution patterns is investigated also by substitution of a methoxy group. Methoxy substitution change the basic character from cis isomer to trans.

[1] Beilstein J. Org. Chem. 2012, 8, 1003.[2] J. Phys. Chem. A 2009, 113, 13144.



Two-dimensional electronic spectroscopy of molecules in helium nanodroplet isolation

U. Bangert, L. Bruder, M. Binz, D. Uhl, and F. Stienkemeier

Institute of Physics, University of Freiburg, Germany

We present the first two-dimensional electronic spectroscopy (2DES) study of cold molecules (sub Kelvin internal temperature) prepared by helium nanodroplet isolation (HENDI) [1]. In contrast to conventional pump-probe spectroscopy, 2DES reveals coherent and incoherent dynamics in molecular systems, giving detailed insight into system-bath interactions, charge/energy transfer and structural changes [2]. Until now, most 2DES studies were conducted with liquid/solid phase samples, due to their high target density. However, with the recent development of highly sensitive methods, dilute gas phase samples like molecular beams have become accessible. The HENDI technique is of special interest, since it provides internally cold molecules and flexible doping capabilities. The latter is used to synthesize arbitrary molecular complexes.

As a first benchmark system, we studied weakly bound Rb molecules attached to helium nanodroplets. Our results, including dynamics due to molecule-matrix interaction, show the applicability of 2DES to samples with extreme low target densities ($\leq 10^7$ cm⁻³).

The final goal of the project is to study charge transfer in organic complexes. We currently take the next step by investigating our first organic molecule, Phthalocyanine, embedded in helium nanodroplets.





Figure 1

2D-spectrum of Rb_2 and Rb_3 attached to helium nanodroplets at a pump probe delay of 200fs. ω_{τ} and ω_t are the pump and probe frequency respectively. The Rb_2/Rb_3 features are assigned by comparison of the spectral profile along the pump axis to high-resolution absorption spectra.

References

- [1] L. Bruder et al., *Nature Communications* **9**: 4823, (2018).
- [2] F. D. Fuller & J. P. Ogilvie, *Annual Review of Physical Chemistry* **66**, 667–690 (2015).



High intensity effects in two-dimensional electronic spectroscopy

M. Binz¹, U. Bangert¹, L. Bruder¹, F. Landmesser¹, D. Uhl¹, L. Chen², M. F. Gelin², W. Domcke², and F. Stienkemeier¹

¹Institute of Physics, University of Freiburg, Germany ²Chemistry Department, Technical University of Munich, Germany

Usually, two-dimensional electronic spectroscopy (2DES) experiments are performed in the regime where perturbation theory holds and the signal can be described by the third-order polarization. However, to measure nonlinear signals, higher laser intensities are generally of advantage as the signal scales with higher order of the incident light fields. Furthermore, new 2DES schemes have been implemented [1,2] where tight focusing is crucial and thus laser intensities beyond the weak-perturbation limit may be reached. Recently, a theoretical description for 2DES experiments beyond the weak perturbation limit has been reported [3] showing peak shape distortions and phase shifts, which may be exploited to gain additional information but can also lead to artifacts. In collinear 2DES experiments, we explore these high intensity effects in our lab by studying a simple, clean model system comprising of a rubidium atom vapor. By comparing these experiments with simulations, we hope to get a direct intuition of intensity effects in 2D spectroscopy.

References

- V. Tiwari, Y. A. Matutes, A. T. Gardiner, T. L. C. Jansen, R. J. Cogdell, and J. P. Ogilvie, Nat Commun 9, 4219 (2018)
- S. Goetz, D. Li, V. Kolb, J. Pflaum, and T. Brixner, Opt. Express 26, 3915-3925 (2018)
- [3] L. Chen, E. Palacino-González, M. F. Gelin, and W. Domcke, J. Chem. Phys. 147, 234104 (2017)



Spectroscopy of oligoacene molecules attached to argon clusters

Matthias Bohlen [1], Moritz Michelbach [1], Rupert Michiels [1], Corey Allen Rice [1], Aaron LaForge [1,2], Frank Stienkemeier [1]

[1] Institute of Physics, University of Freiburg, Freiburg, Germany[2] University of Connecticut, Department of Physics, Storrs, CT, USA

The energy conversion efficiency of solar cells is generally limited by the Shockley-Queisser limit [1]. One way to circumvent this limit is through the use of organic photovoltaics (OPV), where specific charge and energy transfer processes can lead to higher conversion efficiencies. Polyaromatic hydrocarbons such as oligoacene molecules exhibit interesting quantum effects such as singlet fission, triplet-triplet annihilation, or superradiance and make promising candidates for OPV applications. Recently, anthracene, tetracene and pentacene molecules attached to the surface of neon clusters have been shown to provide interesting model systems for detailed studies of such effects [2]. We have extended these measurements to the spectroscopy of oligoacenes deposited to the surface of argon clusters. For this purpose, we developed a liquid-nitrogen-source to cover a wide temperature range for the production of argon clusters. First results show tremendous disturbances of the fluorescence signals due to Rayleigh-scattering from argon clusters, which necessitates an extensive single-shot analysis to extract fluorescence information. First results indicate collective lifetime reducing effects for pentacene attached to argon clusters, exhibiting clear similarities to previous investigations on neon matrices.

[1] W. Shockley and H. J. Queisser, J Appl Phys **32**, 510 (1961)
[2] S. Izadnia et al., J. Phys. Chem. Lett. **8**, 2068 (2017)



Time-resolved imaging of the dynamics of free metal clusters and nanocrystals

Simon Dold

Institute of Physics, University of Freiburg, Germany



X-Ray diffraction has been proven a viable tool to determine the 3D structure of metal clusters [1]. The temporal resolution of this technique is only limited by the duration of the FEL x-ray puls (≈100fs) which enables imaging ultrafast processes. Such diffraction experiments on metal clusters in gas-phase are highly demanding as they require high intensities of both, clusters and X-ray beam to vanquish low cross-sections for scattering processes. To overcome this difficulties a carefully taylored source for clusters was set up. I will report on the results of our efforts to utilize ultrashort X-Ray pulses as a tool to resolve ultrafast processes in nanoscale systems by imaging structure in a time-dependent manner.

References

[1] Ingo Barke, et al. The 3d-architecture of individual free silver nanoparticles captured by x-ray scattering. *Nature communications*, 6, 2015.



100 kHz Attosecond Pulse Source for Coincidence Spectroscopy

Dominik Ertel^{1*}, Anna-Lena Jäger¹, Samuel Kellerer¹, Matteo Moioli¹, Hamed Ahmadi¹, Fabio Frassetto², Luca Poletto² and Giuseppe Sansone¹

¹Institute of Physics, University of Freiburg, Freiburg, Germany ²CNR-Institute of Photonics and Nanotechnologies (CNR-IFN), Padova, Italy *dominik.ertel@physik.uni-freiburg.de

Attosecond coincidence spectroscopy builds a powerful tool to study ultrafast phenomena in atoms and molecules [1, 2, 3]. Here, we present an attosecond extreme ultraviolet (XUV) pulse source with high repetition rates up to 100 kHz, developed for photoelectron/-ion coincidence spectroscopy. Near-infrared (NIR) laser pulses with up to 400 μ J pulse energy and a pulse duration of around 300 fs are temporally compressed using a gas-filled hollow-core fiber [4, 5]. Afterwards, we use these pulses to generate trains of attosecond pulses via high-order harmonic generation in noble gases. RABITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) based coincidence pump-probe spectroscopy is performed using a reaction microscope. The long acquisition measurements require an ultrastable control of the delay between pump (XUV) and probe (NIR). A novel non-interferometric delay line enables intrinsic delay synchronization within a few attoseconds over several hours. Our coincidence spectroscopy setup will enable the investigation of electronic and coupled electronic-nuclear dynamics. This could shed new light on the first instants of photoionization and subsequent nuclear dynamics in small-sized molecules.

References

- [1] F. Krausz et al., Rev. Mod. Phys. **81**, 163 (2009).
- [2] J. Vos et al., Science, **360**, 1326 (2018).
- [3] L. Cattaneo et al., Nature Physics, **14**, 733 (2018).
- [4] M. Nisoli et al., Appl. Phys. Lett. **68**, 2793 (1996).
- [5] J. Beetar et al., J. Opt. Soc. Am. B **36**, A33 (2019).



Singlet fission in Pentacene dimers on top of Argon cluster surfaces

Selmane Ferchane

Institute of Physics, University of Freiburg, Germany

Singlet fission (SF) is a spontaneous photoexcited splitting phenomenon in which an organic chromophore in an excited singlet state shares its energy with a neighboring chromophore in ground-state converting into a pair of triplet excited states. Singlet fission is a process that holds a potential to supplant the traditional solar energy conversion scheme (Shockley–Queisser limit) as a great promise for future photon-to-current conversion solar energy organic materials with high efficiencies. Although, the singlet fission process has been known since decades, as the phenomena remains relatively obscure about how the mechanism is undergoing to this day.

In order to get more insight into these processes, we investigate pentacene dimer chromophores on top of Argon cluster within density functional theory (DFT). Similar systems are experimentally studied in the Stienkemeier group. In the first step, a number of Pentacene dimers with different spatial configurations are studied in space and on top of the Argon cluster surfaces. We calculate binding energies, most favorable positions with lowest ground state energy and other ground state properties prior to the investigation of singlet fission phenomenon itself.



Rotational motion of carbon disulfide in helium nanodroplets

Jordan Fordyce, Ian MacPhail-Bartley, Frank Stienkemeier*, and Valery Milner

Department of Physics and Astronomy, University of British Columbia, Vancouver, Canada *Physikalisches Institut, Universität Freiburg, Freiburg, Germany

Nanodroplets ideally capture single carbon disulfide molecules and are then exposed to two laser beams to study the behaviour of fast rotating molecules immersed in superfluid helium. The first beam excites the molecule to some rotational state and the second beam, a femtosecond "probe", ionizes the molecules. The first "pump" beam can be a femtosecond "kick" pulse or a laser pulse whose linear polarization undergoes accelerated rotation around the direction of the laser beam , called an "optical centrifuge", that excites molecules to extreme rotational states. The recorded velocity map ion image is analyzed to study the dynamics of the rotational state by quantifying the degree of confinement of the molecules to the rotational plane.

Driving a rigid rotor to an extreme rotational state confines the molecules to the plane almost perfectly, but the bonds are actually soft and can stretch. Molecules like nitrogen spin like rigid rotors, but molecules like carbon disulfide have their rotational motion strongly affected by centrifugal distortion. For carbon disulfide this type of behaviour occurs at high rotational frequencies; around 5.5 THz 1, even though the optical centrifuge can spin up to 10 THz. A helium droplet may couple to a superrotor and prevent rotational motion by amplifying the centrifugal distortion experienced by the molecule 2, lowering the final rotational frequency.



It has been demonstrated that for some molecules the rotational dynamics in helium are the same when excited by a kick pulse but others are impacted by the centrifugal distortion. This new development could explain some of the challenges exciting molecules with the optical centrifuge. Moving forward, a kick pulse can be used as an indication of the expected degree of planar confinement for candidate molecules to be driven by the centrifuge. The first step in doing this is to benchmark our operating point using carbon disulfide.

1: A. Milner, et al., J. Chem. Phys., **147**, 124202 (2017) 2: I. Cherepanov, et al., arXiv:1906.12238 (2019)



The Auger Effect in Dispersing and Absorbing Environments

J. Franz¹, R. Bennett^{1,2}, and S. Y. Buhmann^{1,2}

¹University of Freiburg, Germany ²Freiburg Institute for Advanced Studies, Germany

The Auger effect is the radiationless decay of an inner-shell ionised atom. In this process, the atom relaxes by filling the inner vacancy with an outer shell electron, but instead of releasing the excess energy in form of a photon (spontaneous decay) the energy is reabsorbed by another electron belonging to the same atom. This effect finds many applications: for example in Auger electron spectroscopy, it is used to study material properties of surfaces. It is well established that dielectric environments can have a significant impact on spontaneous decay (Purcell effect) [1] as well as interatomic energy transfer rates [2]. We present a general expression for the intra-atomic Auger process in the presence of dielectric environments that can be used both as a new starting point for ab initio quantum chemistry, or within the quantum-optical formalism of macroscopic quantum electrodynamics in order to circumvent complex numerics. Within our description, the decay rate can be given in analytical form for some simple environments. We compare Auger decay with a competing process known as interatomic Coulombic decay, focussing on their behaviour in the presence of surfaces.

REFERENCES

- 1. Purcell, E. M., Proc. Am. Phys. Soc., Vol. 69, 674, 1946.
- 2. Hemmerich, J. L., R. Bennett, and S. Y. Buhmann, Nature Commun., Vol. 9, 2934, 2018.



Quantum state dependent chemistry of ultra-cold 6Li2 dimers

Erik Frieling

Department of Physics and Astronomy, The University of British Columbia, Vancouver, CA

Reactive and inelastic collisions of ultra-cold molecules has generally been observed to follow a universal rate law described by the quantum Langevin model. The salient feature of this law is an independence of the reaction probability from the short-range physics of the interaction. We report on reactive and inelastic collisions of 6Li2 dimers in several ro-vibrational states of the a(13Σ +u) potential. While the v = 0, 5, 8 are observed to decay at the universal limit, decay of the $|v = 9, N = 0\rangle$ state exhibits a deviation from universality, opening up the possibility of using external magnetic fields to tune the reaction rate.



<u>Many-body effects in cold molecules using phase-modulated</u> <u>two-dimensional coherent spectroscopy</u>

Friedemann Landmesser, Ulrich Bangert, Lukas Bruder, Marcel Binz, Daniel Uhl and Frank Stienkemeier

Institute of Physics, University of Freiburg, Germany

Many-body guantum states are considered to play a crucial role in atomic and molecular systems with respect to dissipation as well as excitation and energy transfer processes (cf. quantum efficiencies of biological light harvesting complexes [1]). We aim to investigate collective effects in organic molecules by multiple-quantum coherence experiments where multiphoton processes can be separated from one-photon transitions and can be assigned to specific particle numbers [2,3]. In a first step, we will adapt a detection scheme based on phase-modulated two-dimensional coherent spectroscopy which was already used to investigate multi-atom Dicke states in potassium vapor [2,4]. Measurements on a rubidium vapor will serve as a benchmark. To apply the detection scheme to organic molecular systems, we will adapt our helium nanodroplet source to produce solid rare gas clusters, that can be doped with hundreds of organic molecules. The cluster surface acts as a well-defined, cold environment [5]. In lifetime measurements, collective effects of weakly interacting organic molecules have already been identified at increasing doping densities [5].

- F. Fassioli, R. Dinshaw, P. C. Arpin, and G. D. Scholes, J. Royal Soc. Interface 11, 20130901 (2014).
- [2] S. Yu et al., Opt. Lett. 44, 2795 (2019).
- [3] L. Bruder, M. Binz, and F. Stienkemeier, Phys. Rev. A 92, 053412 (2015).
- [4] L. Bruder et al., Phys. Chem. Chem. Phys. 21, 2276 (2019).
- [5] S. Izadnia et al., J. Phys. Chem. Lett. 8, 2068 (2017).



Spectroscopy of pentacene dimers in rare gas matrices

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Oligoacene molecules such as pentacene are promising candidates for organic solar cells which can achieve photon-to-current conversion efficiencies beyond the 30% Shockley-Queisser¹ limit. The underlying process to create multiple charge carriers from a single photon is called singlet fission where an excited molecule can partially transfer energy to a neighboring ground state molecule, and thereby create a correlated triplet pair². This conversion process influences the excitation lifetime of the excited molecule and can be measured via time resolved laser induced fluorescence³.

A detailed study of this process needs high temporal resolution and a wellknown intermolecular distance. To use the advantages of rare gas spectroscopy we need to specify the doping conditions and the size distribution of the cluster beam. Thus, we investigated the cluster sizes with various methods such as the so-called titration method or Rayleigh-scattering to aid a better understanding of collective effects of chromophores attached to these clusters.

A different approach is the deposition of bis-TIPS-pentacene as a "macromolecule ", accommodating two pentacene molecules in a well-defined distance as a model system⁵. With this we want to investigate an intramolecular singlet fission process in a spectral and time resolved manner.

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Towards Cold Chiral Radicals

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The origins of homochirality, the existence of biological molecules as specifically one enantiomer in living systems, have been a subject of debate in recent years. It has been shown that the presence of enantiomeric excess can be amplified through fundamental chemical processes, leading to a homochiral system. One theory that can explain this enantiomeric excess is the difference in energy between enantiomers of chiral molecules due to parity violation. For most chiral molecules, this energy difference is on the order of mHz to Hz. A recent study by Suzuki et al. has shown that chiral radicals have enhanced parity violation energy differences up to kHz. In combination with molecular slowing techniques, this energy difference is feasible to measure experimentally. Zeeman deceleration has been proven effective for slowing and trapping of radicals, and is therefore a suitable technique for the trapping of chiral radicals. We will characterize the chiral radical production by photolysis and electric discharge in combination with matrix isolation spectroscopy in solid parahydrogen. Our initial radical production studies as well as matrix-isolated spectroscopic studies of the parent molecule used for radical production will be presented.



<u>Fragmentation channels of pentacene and pentacene embedded in</u> <u>helium nanodroplets upon electron impact ionization</u>

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The isolation of atoms, molecules and molecular complexes in rare-gas cluster beams, and particularly in helium nanodroplets, offers a versatile method for the study of energetic and dynamical properties of these systems, because the cluster environment provides low temperature conditions with minimal perturbation by the environment[1]. The interaction of the molecule with the cold He environment leads to a strong cooling of the molecular degrees of freedom. In that case, He nanodroplets may freeze some of the fragmentation channels observed in an effusive beam, especially those which appear after subsequent fragmentation of short-lived decomposition intermediates[2], [3], [4]. Moreover, depending on whether the helium nanodroplets are formed in the subcritical or supercritical regime, it is expected that the charge-transfer mechanism to the dopant competes with the fragmentation of the helium clusters [5], [6]. In addition to charge-transfer ionization, Penning ionization is also playing a role in the observation of multiply-ionized parent ions [7].

In this contribution, preliminary results of pentacene fragmentation pathways in the absence and in the presence of He nanodroplets are presented. In addition to that, first characterization measurements of the He droplet source will be discussed.



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Quantum-state-controlled Penning collisions between lithium atoms and metastable helium atoms

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Our work is aimed at understanding the mechanistic details of reactive collisions and at controlling the outcome of chemical reactions. In our experiment, we study quantum-state-controlled Penning collisions between lithium atoms (Li) and metastable helium atoms (He*) at various collision energies, e.g., to study the influence of electron-spin polarization on the reaction rate. For this, we have combined a supersonic beam source for He* atoms with a magneto-optical trap (MOT) for Li atoms.¹ Our initial investigations have shown that the reaction rate dramatically depends on the initial electronic states of the reaction partners. In order to get full quantum-state control of the reaction partners, the Li atoms are optically pumped into selected electronic hyperfine and magnetic substates. Additionally, we produce a pure beam of $He(2^{3}S_{1})$ by the optical depletion of the $He(2^{1}S_{0})$ state using a novel excitation scheme.² In this contribution, I will give an overview over the different methods used to achieve guantum-state control of the reaction partners and I will discuss experimental results that show the dependence of the reaction rate on the initial electronic state of both He* and Li.

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<u>Thermometry of a single trapped ion immersed in an ultracold</u> <u>atomic gas</u>

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Atom-ion mixtures at ultracold temperatures could provide an interesting system to investigate quantum phenomena of long-range attractive potentials [1,2]. Reaching temperatures below the s-wave threshold, at which only a single partial wave contributes to the scattering process, is of special interest for experimental control of the interaction. This regime could be reached by sympathetic cooling of a heavy ion (¹³⁸Ba⁺), trapped in a radio-frequency trap, with a light coolant (⁶Li) [3].

In this talk we will present and characterize our method to measure the temperature of a single trapped ion. As the intended application, we will show first results that demonstrate sympathetic cooling of a barium ion in a cloud of lithium at a temperature of approximately 50μ K.

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Development of a software based lock-in amplifier for phase-modulated spectroscopy

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Two-dimensional electronic spectroscopy (2DES) is a powerful tool to study coherences and correlations on ultrafast time scales. We implemented a 2DES setup combined with a doped helium droplet beam in our lab. However, the target density in doped helium droplet beams is several orders of magnitudes lower than in bulk condensed phase samples. We adapted a phase modulation technique [1] that introduces a lock-in detection to overcome this issue.

To further improve our detection scheme, we developed a software based lockin amplifier. This offers the possibility to demodulate our signal at different harmonics and thus gives us the advantage to observe any desired separate multiphoton processes in the pre-analysis. The algorithm is mainly based on a digital phase locked loop to detect the phase and frequency of the incoming reference signal. Hence, it is possible to demodulate the signal for different harmonics by conserving the phase information. It also provides a much higher flexibility with our measurements such as simultaneous detection with custom demodulator and filters algorithms or individually optimized acquisition times for specific applications.

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Production of Ultracold Lithium Triplet Dimers by STIRAP

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Ultra-cold atomic ensembles have enabled experimental studies of few and many-body quantum phenomena including topological insulators, many-body pairing phenomena, and superuidity. The use of ultra-cold molecular ensembles is expected to provide access to even richer phenomena than atoms due to their complex internal structure. Cold molecules are also key to studying and understanding chemistry near T=0 where reactions are dominated by quantum e_ects. Motivated by this, we have developed an experimental apparatus that can produce Li dimers from laser-cooled Li atoms and, eventually, LiRb molecules from Li+Rb atomic ensembles. Beginning with a gas of Li atoms, we form loosely bound Feshbach dimers by evaporation near a Feshbach resonance. We then manipulate the internal state of these molecules, transferring them into deeply bound ro-vibrational levels of the $a(1^3\Sigma^+_u)$ triplet potential using stimulated Raman adiabatic passage (STIRAP). Having the possibility to transfer an ultra-cold molecular ensemble to di_erent quantum states immediately opens the window for studies in ultra-cold chemistry.



<u>Trapping polyatomic molecules using Microwaves inside a</u> <u>superconducting cavity</u>

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The trapping of molecules in microwave fields was proposed many years ago [1], but it has not been realized so far. In the last several years we have been investigating the manipulation and trapping of polyatomic molecules using strong microwave field created in superconducting microwave cavity [2]. Here we report our latest progress on the possibility of trapping molecules in a microwave trap we are developing.

Here, microwave radiations close to the inversion doublet splitting in paraammonia with quantum numbers, |J,K >= |1, 1 > are applied to a superconducting cavity at a temperature of 3 K. A superconducting cavity with a high-quality factor will provide a deep potential to manipulate motions of polar molecules. We will discuss the characterization of the cavity by measuring the quality factor at various temperature from 298 K to 3 K. We will also discuss the AC stark shifts of NH3 molecules under the fields, which we are currently working on.

Our discussion is based on using this cavity for trapping stark decelerated NH3 molecules. The successful demonstration of trapping NH3 molecules inside a microwave trap will open new pathways to cool polyatomic molecules below 1 mK via sympathetic cooling. Development of microwave trap will enable us to trap the ground state of polar polyatomic molecules. These ground state molecules are best suited for sympathetic cooling with ultra-cold atomic gases due to their low losses during the thermalization process.



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Building a portable, cold-atom pressure standard

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Vacuum measurement plays a central role in a wide range of scientific and industrial applications including residual gas analysis, semi-conductor device manufacture, and atmospheric modelling. Remarkably, no primary pressure standard existed for the high (HV) and ultra-high vacuum (UHV) regime (below 10[^]–7 Pa) until recently. In 2018, a UBC and BCIT collaboration succeeded in producing the first primary pressure standard for HV and UHV. Based on a coldatom sensor, this new technique for particle detection requires no calibration and relies on immutable laws of nature - specifically, the interaction potentials between atoms and molecules. The existing cold-atom standard is immobile and the size of several refrigerators; to see wide use and to promote worldwide adoption, an apparatus must be shippable, and usable in non-ideal conditions by non-experts. To realize this, we are assembling a miniaturized version of the apparatus that will be robust to vibrations, mechanical shock, and temperature changes. We will also investigate methods of automatically operating and recalibrating the apparatus. This device will allow us to disseminate the cold-atom pressure standard worldwide starting with a plan to compare the cold atom primary standard with an existing secondary pressure standard (based on orifice-flow) at the National Institute of Standards and Technology. This work will also lay the groundwork for the construction of commercial absolute pressure standards.



Preparing and probing an ultracold fermi gas for atom-ion experiments

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The interplay of ultracold atoms and ions has gained interest in the atomic physics community [1,2], due to its wide applications in quantum simulations of solid state systems [3] as well as in quantum computing [4]. In order to implement a reliable atom-ion platform, it is necessary to prepare the mixture at ultracold temperatures, where only few partial-waves contribute to the scattering process. Choosing a proper atom-ion mixture with high mass imbalance (as ⁶Li and ¹³⁸Ba⁺) has recently been proposed to be a promising candidate for reaching the quantum limit [5].

In this talk our setup creating 10^4 ⁶Li-atoms at a temperature of 50μ K will be presented. This cold fermi gas is realized by evaporatively cooling a thermal gas in a crossed optical dipole trap at high magnetic fields. The implementation of these magnetic fields as well as our detection scheme, high field absorption imaging, will be introduced.

As an outlook we discuss our first measurements between a single 138 Ba⁺ ion immersed in a cold cloud of Li atoms. Hereby we observe sympathetic cooling into the few partial-wave regime.

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Initial-state dependence of the long-time dynamics in a spin-boson system

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Thermalization of a system refers to the process of approaching a unique steady state that depends only on a few macroscopic observables. Triggered by the development of new experimental tools, for example in cold atom or trapped ion experiments, the question how an isolated quantum system can equilibrate to such a state under its own dynamics has recently received renewed interest. Various numerical studies suggest that subsystems of isolated quantum systems can equilibrate under unitary time evolution. This relaxation is caused by the remainder of the system acting as an effective bath. However, as the hybridization between the subsystem and the remainder increases this simple picture breaks down and the influence of the remainder on the subsystem can change qualitatively.

Inspired by a recent experiment with trapped-ions [1], we investigate a special type of a spin-boson model. Employing the multilayer multiconfiguration time dependent Hartree method [2], we simulate the dynamics of the system in a numerically exact way. As a function of the spin-boson coupling, we observe a qualitative change of the dynamics of the spin, induced by the coupling to the bosonic modes, ranging from the usual relaxation dynamics for small couplings to non-stationary long-time states of the spin that depend on its initial state.

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<u>Tracking Attosecond Electronic Coherences Using Phase-Manipulated</u> <u>Extreme Ultraviolet Pulses</u>

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Experiments utilising advanced coherent control and nonlinear spectroscopy schemes are powerful tools that allow the study and control of the dynamics of atomic and molecular systems. The advent of novel light sources in the extreme ultraviolet (XUV) region paved the way for the extension of these tools to the XUV, potentially allowing similar experiments with an unprecedented spatial and temporal resolution. However, this promise has not yet been honoured due to the experimental difficulty of creating XUV pulse trains with precisely controlled timing and phase properties.

In this work we use an XUV pulse pair to measure directly in the time domain the coherent evolution and dephasing of electronic XUV wave packets. We demonstrate this in helium and for an autoionizing resonance in argon (Fig. 1). Two collinear XUV pulses were created at the FERMI free-electron laser (FEL) operated in double seed-pulse mode [1]. We controlled the relative phase and delay of the pulse pair independently by acting only on the fundamental ultraviolet seed pulse [2]. Imparting a shot-to-shot phase modulation lead to modulated photoion/electron yields [3]. Subsequent lock-in demodulation



shows a particularly high sensitivity and allows the isolation of different harmonic contributions in the total signal. This phase manipulation technique facilitates a wide range of nonlinear spectroscopy as well as coherent control schemes, and is universally applicable to the whole wavelength range accessible with seeded FELs. In addition, it can be combined with tabletop High Harmonic Generation sources.



Fig. 1 a) Excitation scheme for preparation and probing of the Ar 3s-6p inner subshell-valence coherence **b)** Demodulated ion signal (real part, amplitude, phase) measured in the rotating frame. **c)** Fourier transform. The characteristic Fano line shape is recovered due to phase sensitive detection



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ξ	4th IRTG CoCo Summer School Cold Molecules and their Interactions Theoretical and Experimental Methods 29th July - 3rd August 2019, Kelowna, Canada UBC Okanagan Campus, Kelowna					
	Monday 29th July 2019	Tuesday 30th July 2019	Wednesday 31st July 2019	Thursday 1st August 2019	Friday 2nd August 2019	Saturday 3rd August 2019
09:00		Sergey Bokarev	Michael Tarbutt	Gary Douberly	Olivier Dulieu	James P. Shaffer
		Electronic structure theory for photophysics of isolated molecules	Formation and application of cold and ultracold molecules	Aggregation in Helium Droplets	Theory of cold molecules	Rydberg Molecules, Interactions and Applications
10:30]	Break	Break	Break	Break	Break
11:00		Sergey Bokarev	Michael Tarbutt	Gary Douberly	Olivier Dulieu	James P. Shaffer
		Electronic structure theory for photophysics of isolated molecules	Formation and application of cold and ultracold molecules	Aggregation in Helium Droplets	Theory of cold molecules	Rydberg Molecules, Interactions and Applications
12:30		Lunch break	Lunch break	Lunch break	Lunch break	Lunch break
14:30		Sergey Bokarev	Excursion	Gary Douberly	Olivier Dulieu	James P. Shaffer
	Arrival	Electronic structure theory for photophysics of isolated molecules		Aggregation in Helium Droplets	Theory of cold molecules	Rydberg Molecules, Interactions and Applications
16:00	Registration	Break	Hiking	Break	Break	Break
16:30	Welcome & Introduction	Michael Tarbutt Formation and application of cold and ultracold molecules	Visiting a Winery &	Poster Karaoke Session I	Poster Karaoke Session II	Closing Session
18:00			Dinner			
19:30	Dinner	Dinner		Dinner	Dinner	