Abstracts & Topics for Mildred Dresselhaus Jubilee 2024

Jubilee Talks Day 1: 14.3.2024

Prof. Dr. Alicia Palacios, Universidad Autónoma de Madrid

"Time Management" & "A glance of recent applications to explore attosecond electron dynamics"

Jobs in research and academia have shown to present high levels of stress coming from multiple sources. One of the main work-related stress factors is the need of multitasking, for which, the ability of an effective time management can make a difference. The first part of the talk will provide a review on available tools for efficient time management and on planning skills in the academic environment.

As an hybrid talk, in the second part, a brief summary of recent applications on attosecond pump-probe techniques to access ultrafast electron phenomena in atoms and molecules in gas phase will be presented. Ongoing progress on the implementation and developing of new theoretical tools to describe these processes will be briefly discussed.

Prof. Dr. Rosario Ferez, Universidad de Granada

Ultralong-range Rydberg Molecules: Electronic Structure and Rydberg blockade

R. González-Férez

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The exotic properties of Rydberg atoms make them unique probes of their environments. In hybrid systems, they form ultralong-range molecules when combined with ground-state atoms [1, 2], ions [3], or polar molecules [4,5], which inherit these exciting properties. When the diatomic polar molecule is immersed into the wave function of the excited atom, the anisotropic scattering of the Rydberg electron from the permanent electric dipole moment of the dimer is responsible for the binding mechanism in these Rydberg molecules [4, 5]. In this work, we explore the electronic structure and main properties of these exotic ultralong-range molecules, which are formed by a Rydberg atom interacting with RbCs. Our focus is the regime where the charge-dipole interaction of the Rydberg electron with the diatomic polar molecule induces a coupling between the quantum defect Rydberg states and the nearest degenerate hydrogenic manifold [6]. We present adiabatic electronic states evolving from the Rydberg degenerate manifold and from the quantum defect states, and analyze the non-adiabatic coupling between these potentials, and decay rates and formation rates. In addition, we present the first experimental demonstration of the Rydberg blockade due to the charge-dipole interaction between a single Rb atom and a single RbCs molecule. The atom and molecule are confined in optical tweezers,

which are used to control their relative distance. For a separation of 310 nm, the charge-dipole interaction between the Rydberg electron and atomic core with the dipole moment of RbCs provokes the blockade of the transition to the Rb Rydberg state. The observed excitation dynamics are in excellent agreement with the theoretical results obtained using the electronic structure of the Rydberg Molecule Rb-RbCs [7].

References

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[2] V. Bendkowsky et al, Phys. Rev. Lett. 105, 163201 (2010)

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[6] R. González-Férez, et al, J. Phys. B: At. Mol. Opt. Phys. 53, 074002 (2020).

[7] A. Guttridge et al, Phys. Rev. Lett. Phys. Rev. Lett. 131, 013401 (2023).

Prof. Dr. Roseanne Sension, University of Michigan

Watching Molecules in Action: Cobalamins as a case study

The fate of a photoactive molecule is determined by the electronic and structural rearrangements that follow excitation. Femtosecond X-ray free electron lasers (XFELs) have made it possible to use X-ray absorption spectroscopy to probe changes in electronic configuration and atomic structure as a function of time, beginning from the initial excited state. Both 'movies' of coherent or ballistic motion and 'snaphots' of local minima or kinetic intermediates are possible. Polarization anisotropy, long exploited in ultrafast optical measurements, permits decomposition of the X-ray transient difference signal into contributions along the direction parallel to the transition dipole initially pumped, and perpendicular to this transition dipole. This decomposition allows the analysis of asymmetric sequential structural changes of photoexcited molecules in isotropic solution. We have used femtosecond X-ray absorption near edge structure (XANES) at the Co K-edge to characterize the excited state dynamics of cobalamins, B_{12} coenzymes and analogues at the Linac Coherent Light Source (LCLS) in California. Femtosecond X-ray Emission spectroscopy (XES) and Co-L edge absorption measurements at the Eupopean XFEL provide additional insight into the electronic evolution coupled to structural rearrangement. In addition to presenting the experimental results and future potential of ultrafast X-ray spectroscopy, the talk will also focus on the skills and organization required for running experiments at these facilities.

Prof. Dr. Prineha Narang, UCLA

Out of equilibrium control of quantum matter

Prof. Dr. Ruth Signorell, ETH Zurich

Aerosol particles: Photochemistry, formation, and imaging

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Photochemical processes have been identified as the main causes of degradation and oxidation of matter in atmospheric aerosol particles. When light interacts with an aerosol particle, the light intensity can be greatly amplified inside the particle as the latter acts as a light-amplifying cavity. These optical confinement effects result in an acceleration of photochemical reactions in aerosol particles compared with reactions in extended condensed matter. We have studied and quantified the acceleration of in-particle photochemistry using photoacoustic spectroscopy [1] and X-ray spectro-microscopic imaging of single aerosol particles [2].

If time permits, I will briefly explain how we investigate the very first steps of aerosol formation, and how we use photoelectron imaging and coherent diffraction imaging for aerosol characterization.

1. J.W. Cremer, K.M. Thaler, C. Haisch, R. Signorell, „Photoacoustics of single laser-trapped nanodroplets for the direct observation of nanofocusing in aerosol photokinectics", *Nat. Commun.,* 7, 10941 (2016)

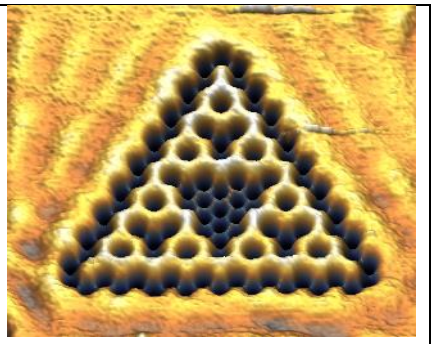
2. P.C. Arroyo, G. David, P.A. Alpert, E.A. Parmentier, M. Ammann and R. Signorell, „Amplification of light within aerosol particles accelerates in-particle photochemistry", *Science*, 376, 293-296 (2022).

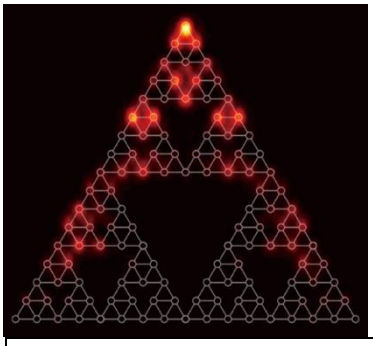
Prof. Dr. Cristiane Morais Smith

Institute for Theoretical Physics, Utrecht University, The Netherlands

Quantum Fractals

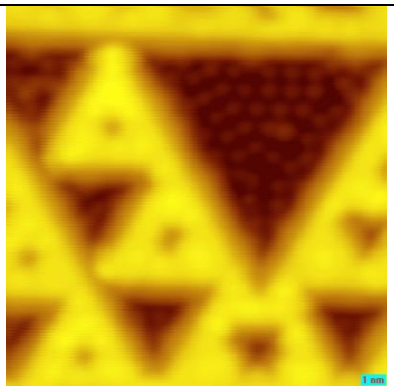
The human fascination for fractals is very ancient, but it was only in the last century that mathematicians classified these structures. In the 80's, the foundational work of Mandelbrot triggered enormous activity in the field. The focus was on understanding classical fractals.

This century, the task is to understand quantum fractals. In 2019, we realized a Sierpinski gasket using a scanning tunneling microscope to pattern adsorbates on top of Cu(111) and showed that the wavefunction describing electrons in a Sierpinski gasket fractal has the Hausdorff dimension d = 1.58 [1]. However, STM techniques can only describe equilibrium properties.	



In 2021, we went a step beyond and using photonics experiments we unveiled the quantum dynamics in fractals. By injecting photons in waveguide arrays arranged in a fractal shape, we were able to follow their motion and understand their quantum dynamics with unprecedented detail. We built 3 types of fractal structures to reveal also the influence of geometry [2].

Recently, we investigated topological effects in self-formed fractals of Bi on InSb [3]. In these systems, the spin-orbit coupling is very strong, thus leading to a quantum spin Hall effect. Muffin-tin calculations indeed reveal topological corner and edge modes in these fractal structures, in agreement with experimental observations.



[1] S.N. Kempkes, M.R. Slot, S.E. Freeney, S.J.M. Zevenhuizen, D. Vanmaekelbergh, I. Swart, and C. Morais Smith, *"Design and characterization of electronic fractals*", Nature Physics 15, 127(2019) [see also 15 years of Nature Physics, Nature Physics 16, 999 (2020)].

[2] X.-Y. Xu, X.-W. Wang, D.-Y. Chen, C. Morais Smith, and X.-M. Jin, "*Quantum transport in fractal networks*," Nature Photonics 15, 703 (2021).

[3] R. Canyellas, Chen Liu, R. Arouca, L. Eek, G. Wang, Yin Yin, D. Guan, Y. Li, S. Wang. H. Zheng, Canhua Liu, Jinfeng Jia, and C. Morais Smith, "*Topological corner and edge states in Bi fractals on InSb*," ArXiv: 2309.09860 (2023).

Prof. Dr. Olga Smirnova

Ultrafast molecular chirality: a topological connection

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An object is said to be chiral if it cannot be superimposed on its mirror image by any rotation. The two mirror images of the same chiral molecule are called enantiomers and are often referred to as “left”- and “right”-handed. While the physical properties of the two enantiomers of the same chiral molecule are nearly identical, the geometric property of chirality leads to vastly different chemical properties of the two enantiomers. The need for rigorous selection of a specific enantiomer, a now standard requirement in drug design, is one of the many reasons behind the ever-greater need for improving sensitivity of chiral sensing.

Yet, standard optical methods of chiral detection still use the same principles as the method discovered by Louis Pasteur in the XIX century: the linear interaction between chiral molecules and light, which becomes chiral-sensitive due to the magnetic field component of the light wave.

Ultrafast non-linear spectroscopies promise to increase the enantio-sensitive signal by three orders of magnitude [1] by removing the need to rely on the interaction with the magnetic field component of light. The second important feature of non-linear light-matter interactions is the opportunity to imprint topological properties of light on matter, presenting an opportunity to achieve topologically robust enantio-sensitive observables.

I will describe our very recent results [2,3] on marrying chiral and topological properties in ultrafast electronic response of chiral molecules in the gas phase, enabling highly efficient and robust chiral observables. I will present two vignettes where topological connection appears in optical or electronic chiral response:

(i) **Chiral topological light**: a new concept enabling chiral-sensitive and topologically robust properties of high harmonic emission, generated by such light in chiral molecular gases [2]
(ii) **Temporal geometry**: a concept encompassing the emergence of geometric fields in electronic response of chiral molecules
(iii) **Enantio-sensitive exceptional points**: chiral topology in non-Hermitian chiral systems [3]

References:

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[2] “Chiral topological light for detecting robust enantio-sensitive observables” N Mayer, D Ayuso, M Ivanov, M. Khokhlova, E Pisanty, O Smirnova, arXiv preprint arXiv:2303.10932, 2023

[3] “Enantiosensitive exceptional points” N Mayer, N Moiseyev, O Smirnova, arXiv preprint arXiv:2306.12293, 2023

Day 2, 15.3.2024

Prof. Dr. Elspeth F. Garman, University of Oxford

Radiation damage and metal identification in Structural Biology: why do we care?

Structural biology relies on X-ray crystallography to provide much of the three dimensional information on macromolecules that informs biological function [1]. My group has helped to establish improved methods for macromolecular crystallography (MX) to enable problems not previously accessible to structure solution to be tackled. A notable example has been the development of protocols to cryocool protein crystals prior to diffraction data collection at 100K, reducing the rate of radiation damage (RD) [2] by around a factor of 70 compared to holding the crystal at room temperature. However, even at 100 K, RD is still a limiting problem as it can prevent structure determination and the changes can mislead the experimenter when interpreting the biology of the structure.

Our contributions have included full dose modelling of the diffraction experiment (RADDOSE-3D, www.raddo.se) and RADDOSE-XFEL) to allow data collection optimisation strategies [3,4,5], as well as the recent identification of a single metric, *B*net, by which the level of damage in a single PDB entry can be assessed [6].

Our work also addresses the accurate identification of metal atoms bound to protein structures. Metalloproteins comprise over one-third of proteins, with approximately half of all enzymes requiring metal to function. Identifying the bound metal and its environment is a prerequisite to understanding biological mechanism. However, there are no routine analysis methods with the sensitivity and quantitative accuracy to do this. We have developed microProton Induced X-ray Emission (PIXE) as a tool for quantifying metals in proteins using the known sulphur content (methionines and cysteines) as an internal standard. We have automated this method to permit high throughput analysis of many samples, validating the approach by using it to analyse four distinct sets of 30 proteins identified as metalloproteins in the Protein Data Bank (PDB) [7]. In all four sets, we found that over half of the metals had been misidentified in the deposited structural models. The PDB is a critical resource for researchers worldwide and in 2021 there were on average 1.86 million downloads per day in the US alone, suggesting that over 350,000 models downloaded per day may not contain the correct metal. This has profound implications for those using the models, whose understanding of them may therefore be flawed.

References:

[1] EF Garman (2014) Developments in X-ray Crystallographic Structure Determination of Biological Macromolecules. *Science* 343: 1102-1108

[2] EF Garman. (1999) Cool Data: Quantity and Quality. *Acta Cryst.* D55: 1641-1653.

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[4] OB Zeldin et al & EF Garman (2013) Predicting the X-ray lifetime of protein crystals. PNAS 110: 20551-20556

[5] JL Dickerson, PTN McCubbin & EF Garman (2020) RADDOSE-XFEL: Femtosecond time-resolved dose estimates for macromolecular XFEL experiments. *J.Appl.Cryst*. 53, 549–560

[6] KL Shelley & EF Garman (2022) Quantifying radiation damage in the Protein Data Bank.

Nature Communications 13:1314- 1325

[7] GW Grime et al. & EF Garman (2020) High-Throughput PIXE as an Essential Quantitative Assay for Accurate Metalloprotein Structural Analysis: Development and Application. *J Am Chem Soc.* 142(1):185-197

Prof. Dr. Caterina Vozzi, CNR-IFN Milan

Career Talk on "Learnings gathered throughout a scientific career"

In this talk, I will delve into the realm of ultrafast spectroscopy, presenting a review of my main scientific accomplishments in this field. Through experimental development of laser sources and attosecond technologies, I have contributed to the development of high-order harmonic generation spectroscopy and attosecond science, paving the way for advancements in various applications to molecular spectroscopy and materials science. Beyond the technical achievements, this presentation will also shed light on the soft skills cultivated throughout my career. From communication and networking to problem-solving and critical thinking, my journey in ultrafast spectroscopy has been characterized by continual personal and professional growth. I will share insights into both the fascinating world of ultrafast spectroscopy and the essential soft skills that have helped me succeed in science and prepared me for future challenges.

Prof. Dr. Giulia Fulvia Mancini, University of Pavia - LUXEM

Part 1: Multiscale Ultrafast Microscopy across X-ray tabletop and facility-scale sources

Microscopic imaging is critical for discovery and innovation in science and technology. Throughout history, advances in microscopy have dramatically accelerated advances in other areas of science including materials, biological, nano, and energy sciences, as well as nanoelectronics, data storage, and medicine. To this date, current imaging techniques with soft X-ray/EUV light are nowhere near their fundamental limits in terms of spatial, spectral or temporal resolution. In this talk I will present recent advances in this field with X-ray Free-Electron Lasers (XFELs) and High-Harmonic Generation (HHG) sources.

Part 2: Career Talk on Impostor Syndrome

Prof. Dr. Anna Krylov, USC

Water: A perpetual challenge for theory and experiment alike

Spectroscopy provides a way to see what atoms and electrons are doing; however, in order to translate spectroscopic signals into a molecular story, theoretical modeling is needed. I will discuss recent progress in spectroscopy modeling in the condensed phase and illustrate successes and remaining challenges by examples, focusing on the many computational challenges presented by water itself.

Prof. Dr. Liesbeth Janssen, Eindhoven University of Technology

Ideas, Plans, and Proposals

Developing new research ideas, research plans, and research proposals is essential for the modern scientist, yet these skills receive little attention in formal training programs for PhD students and postdocs. In this talk, I will offer some tips and tricks on how to create new research ideas, how to make a critical selection of your ideas and a subsequent research plan, and, if external funding is required to execute your research plan, how to write a competitive research proposal. Overall, I hope that this talk will help you in navigating the path from nothing to ideas, from ideas to plans, and from plans to proposals.

Keynote:

10 Years of the CUI Mildred Dresselhaus program: what have we learned?

Elspeth F. Garman

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Mildred Dresselhaus (MD), the 'Queen of Carbon', was an inspirational scientist and a truly worthy namesake for this innovative guest scientist program at CUI. The first MD Junior and Senior Guest scientists were selected in 2013 for awards presented at a Hamburg Town Hall ceremony in January 2014. Since then, another 18 of us have had the honour of being MD awardees. In 2019 the first 10 of us participated in a 3-day MD Conference which was both enlightening and challenging, as we heard about the wide range of science being researched and also considered the various issues facing women in STEM.

In this talk I will try to draw together, using input from the other awardees, what lessons have been learned in the last 10 years, and perhaps look into my crystal ball to give some pointers for the future development of the MD program.

In my own field of Crystallography, there is a proud historical record and current practice of outstanding researchers who were/are women, in stark contrast to the field in which I was trained: experimental nuclear structure physics. It is interesting to reflect on why crystallography is a positive outlier in the STEM arena, and also how this can not only be maintained but also further improved. William Henry and William Lawrence Bragg, father and son, contrary to the norm for the times during which they worked (first half of the 20th century), had many women in their research groups (WH Bragg: 10/17 and WL Bragg 4/6 [1]), establishing crystallography as a subject in which women don't just survive, but in which they can excel. Crystallographers trained by the Braggs then became inspiring role models for several more generations of highly successful female researchers.

I have been privileged to know and work with some of these scientists, and in this highly personal account, I will comment on the general attitude and climate towards women in STEM and how I perceive that it is changing but still needs further improvement. I will draw on my experiences during the last 47 years as a scientist who happens to be a woman.

Reference:

[1] Maureen M. Julian, "Women in Crystallography," in *Women of Science: Righting the Record*, ed. G. Kass-Simon and Patricia Farnes (Bloomington: Indiana University Press, 1990), pp. 342

Award Ceremony – Scientific Talks

Junior Awardee Dr. Linyuan Zhao

University of Michigan, USA

Probing and Designing Multipolar Orders in Quantum Materials

Complex electronic charge and magnetic spin textures are popularly present in quantum materials, whose long-range ordered phases often call for multipolar orders to describe. As compared to the electronic and magnetic dipolar orders with the electric and magnetic fields to probe, multipolar orders typically lack readily available fields to couple to and hence are often hidden. At the same time, there has been growing evidence to support the significance of multipolar orders in determining the physical properties of a broad class of materials. In this talk, I will describe our recent efforts in probing multipolar orders in three-dimensional synthetic crystals and designing such states in two-dimensional magnetic moire superlattices.

Senior Awardee Professor Lin X. Chen

Ultrafast functional Structural Dynamics of Photoactive Transition Metal Complexes in Solar Energy Conversion Processes

Chemical Science and Engineering Division, Argonne National Laboratory, USA

Department of Chemistry, Northwestern University, USA

Many photochemical events start from initial light-matter interactions that cause atomic and electronic displacements in the excited states away from the energy minima of their potential energy surfaces. When certain photochemical events, such as bond breaking, intersystem crossing and electron/energy transfer taking place within the periods of key vibrational modes, the excited state energies are determined by the trajectories defined by the actual nuclear movements that may lead to different reaction pathways and outcome. Examples will be given in the work of tracking excited state pathways for transition metal complexes on the time scales from femtosecond to a few picoseconds, such as excited state Cu(I) and Pt(II) dimer complexes. Using fs broadband transient spectroscopy and fs X-ray solution scattering, coherent vibrational wavepacket motions can be examined.

From the time evolution of key vibrational modes in the Pt dimer complexes, particularly the Pt-Pt stretching mode, we mapped out excited state trajectories on potential energy surfaces of Pt-dimer complexes for light conversion, including coherent nuclear motions. These studies were carried out in a series of model platinum dimer complexes in solution and their electron donor acceptor complexes, featuring rich photochemistry and a set of intricate excited state potential energy surfaces on time scale previously unattainable.

For the time evolution of the structural dynamics of Cu(I) complex, the experimental results revealed the Jahn-Teller distortion that transforms the symmetry of the molecule from the D_{2d} in the ground state to D_2 in the triplet metal-to-ligand-charge-transfer (MLCT) state along the two key cooperative coordinates, the Cu-N distances and the angle between the two phenanthroline ligand planes. The detailed structural trajectories have been reproduced by quantum mechanical calculations to map out the actual excited state nuclear motions from the Frank-Condon structure to an intermediate triplet state and finally the triplet MLCT state.