"Bringing Molecules to Attention"

Dudley Herschbach, Harvard University

Bretislav is fond of the title of this talk, crafted by him. It was a major theme in the superb Festschrift [1] celebrating his 60th birthday. There he presented an intriguing autobiography, with engaging vignettes about colleagues as well as synopses of his research, teaching, and historical scholarship. These are updated in his exemplary website [2], including an extensive array of his "favourite aphorisms related to Science." During his Harvard era (1987-2003) Bretislav kept molecules and colleagues busy both in my chemistry lab and John Doyle's physics lab. Here I will mention a few episodes and some later consequences. In 1990, Bretislav enjoyed making molecules behave like pendulums. Previously, to do that with electric fields was deemed not practical for molecules in a gas, tumbling like pinwheels. By now, pendular molecules have had key roles in countless experiments, rendered by means of electric or laser fields. A kindred episode dealt with molecular polarizability, previously disrespected as too puny, yet begat further varieties of pendular states. Some offer prospective qubits for quantum computation, others can serve as catalysts for chemical reactions. On arriving at Harvard, Bretislav took part in a Festschrift [3] for the centennial year of Otto Stern (1888-1969). Since then, Bretislav and colleagues have contributed many papers, books, and symposia enriching History of Science.

[1] Molecular Physics, **111**, 1631-1938 (2013).
[2] www.fhi-berlin.mpg.de/mp/friedrich/
[3] Zeitschrift für Physik D **10**, 109-392.

"The Road from Buffer-gas Cooling to Single Trapped Molecules"

John Doyle, Harvard University

Ultracold molecules are a promising platform for diverse scientific goals, ranging from quantum information and simulation to controlled chemistry and precision measurements of fundamental physics. The rich internal structure of molecules, including vibrations, rotations, and hyperfine interactions, provides many handles for exquisite control. However, the very same structural features that make

molecules so desirable to study also complicate the task of controlling them.
25 years ago at Harvard we started work to trap molecules and study their interactions. This was long before the idea of molecules as qbits had come onto the scene. We began with the simple idea that a very cold cryogenic gas could provide the dissipation to load molecules into a magnetic trap. This led to a series of experiments that included the first magnetic trapping of a molecule and study of collisional properties, in particular spin relaxation. We discovered that magnetic trapping was delicate and that only a few species of molecules would be amenable to our initial approach. This led us to separate the cooling mechanism from the trapping mechanism and gave birth to the buffer-gas beam. Later, this beam turned out to be ideally suited to the laser cooling of molecules, as done in several labs now, including our lab with CaF and SrOH.
Laser cooling and magneto-optical trapping of diatomic molecules from a buffer-gas beam has been realized. In our lab temperatures as low as a few μK have been achieved in long-lived, optically trapped samples of CaF. Photon cycling of these types of molecules allows for high fidelity detection of ultracold molecules and for increases in phase space density allowing for, e.g., the study of ultracold molecular collisions. In this talk, I will outline the path from the early days at Harvard to now a very large field of research on cold, trapped molecules including a path to ultracold polyatomic molecules. I will also present our latest results, which includes the trapping and high-fidelity readout of single molecules in optical tweezers.

“Laser-induced alignment and imaging of molecules embedded in helium nanodroplets”

Henrik Stapelfeldt, University of Aarhus

I will show how laser pulses can align molecules in helium nanodroplets and how the ability to place molecules in advantageous spatial orientations allows structural determination of molecular complexes. The talk will focus on the following topics:
(1) <u>Alignment of molecules with pulses much shorter than the molecular rotational periods.</u> The experimental results show rotational

dynamics that differs completely from that of isolated molecules [1]. Notably, pronounced oscillations in the time-dependent molecular alignment, with no counterpart in gas phase molecules, are observed. Angulon theory identifies the oscillations to originate from the unique rotational structure of molecules in He droplets [2].

(2) Alignment induced by pulses that are turned-on (quasi) adiabatically.

It will be shown how the 0.4 K temperature of the molecules inside the droplets enables unprecedented high degrees of alignment, in either one or three dimensions. The method applies to large, complex molecules and the alignment can be made field-free by rapidly switching off the alignment pulse [3].

(3) Femtosecond-laser-induced Coulomb explosion imaging of the structure of molecular dimers and trimers created inside He droplets.

Results for both small linear molecules, including CS_2 [4] and OCS, and the larger molecule tetracene, are presented. Perspectives for time-resolved imaging of bimolecular reactions and interactions are discussed.

[1] B. Shepperson, A. A. Søndergaard, L. Christiansen, J. Kaczmarczyk, R. E. Zillich, M. Lemeshko, and H. Stapelfeldt, Phys. Rev. Lett. **118**, 203203 (2017).
[2] I. N. Cherepanov, G. Bighin, L. Christiansen, A. V. Jørgensen, R. Schmidt, H. Stapelfeldt, and M. Lemeshko, in preparation (2019).
[3] A. S. Chatterley, C. Schouder, L. Christiansen, B. Shepperson, M. H. Rasmussen, Henrik Stapelfeldt, Nat. Comm. **10**, 133 (2019).
[4] J. D. Pickering, B. Shepperson, B. A. K. Hubschmann, F. Thorning and H. Stapelfeldt, Phys. Rev. Lett. **120**, 1121321 (2018).

"The angulon quasiparticle: from molecules in superfluids to ultrafast magnetism"

Mikhail Lemeshko, Istitute of Science and Technology Austria

Recently we have predicted a new quasiparticle - the angulon - which is formed when a quantum impurity (such as an electron, atom, or molecule) exchanges its orbital angular momentum with a many-particle environment (such as lattice phonons or a Fermi sea) [1,2]. Soon thereafter we obtained strong evidence that angulons are

formed in experiments on molecules trapped inside superfluid helium nanodroplets [3]. The angulon theory thereby provided a simple explanation for experimental data accumulated during the last two decades. Moreover, casting the many-particle problem in terms of angulons amounts to a drastic simplification and allows to tackle previously intractable problems related to quantum dynamics [4].
In this presentation we will introduce the angulon concept and discuss novel physical phenomena [1,5,6] arising from the angular momentum exchange in quantum many-particle systems. We will describe the applications of angulons to modern experiments on quantum impurities and on non-equilibrium magnetism [7].

[1] R. Schmidt, M. Lemeshko, Phys. Rev. Lett. 114, 203001 (2015).
[2] R. Schmidt, M. Lemeshko, Phys. Rev. X 6, 011012 (2016).
[3] M. Lemeshko, Phys. Rev. Lett., 118, 095301 (2017); Viewpoint: Physics 10, 20 (2017).
[4] B. Shepperson, A. A. Sondergaard, L. Christiansen, J. Kaczmarczyk, R. E. Zillich, M. Lemeshko, H. Stapelfeldt, Phys. Rev. Lett. 118, 203203 (2017).
[5] E. Yakaboylu, M. Lemeshko, Phys. Rev. Lett. 118, 085302 (2017).
[6] E. Yakaboylu, A. Deuchert, M. Lemeshko, Phys. Rev. Lett. 119, 235301 (2017).
[7] J.H. Mentink, M.I. Katsnelson, M. Lemeshko, arXiv:1802.01638 (2018).

“Bretislav Friedrich and History of Science”

Mary Jo Nye, Oregon State University

Bretislav Friedrich’s scientific work is complemented by historical research and studies that he began publishing in 1996. This talk focuses on some of the leading themes in his historical work. These themes include biographical essays and articles on Otto Stern, Fritz Haber, Otto Sackur, Michael Polanyi, and Clara Immerwahr, along with histories of physical chemistry, the Fritz Haber Institute, and the development of chemical warfare. As in his scientific research, much of the historical work is collaborative, reconstructing important scientific achievements and historical contexts, while often also addressing issues of scientific ethics and responsibility.