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Special Issue Preface

A Tribute to Alec M. Wodtke

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Photo by Gerard Meijer, December 2022

It is our great pleasure to present this Festschrift of the *Journal of Physical Chemistry* to Alec Wodtke - and our community - on the occasion of Alec's 65th birthday. In his inspiring autobiography penned for this Festschrift, he notes that "Science - like creative art - can be part of a life worth living" - a message to entice the young generation to choose a life in science. In writing his autobiography, Alec did not abide by the rules that this journal has set for this, but he held on to his original version without changing a single word. This is characteristic of Alec and consistent with how we know and appreciate him. In a presentation that Alec gave just over ten years ago in Berlin as part of a ceremony in which he was anointed as Alexander von Humboldt Professor, he touched upon questions of curiosity and the joy of learning, of scientific originality and intellectual leadership. He explained that he saw his views on these matters aptly expressed in the poem by Robert Frost, entitled "The Road Not Taken", which he then recited in full. The poem's last three lines read:

> *Two roads diverged in a wood, and I - I took the one less traveled by, And that has made all the difference.*

Throughout his career, Alec has made a difference by tackling a variety of important scientific problems in physical chemistry and chemical physics, for which he often specifically designed new instrumentation as well as exploited novel detection and analysis schemes, i.e., by taking roads less traveled or not traveled at all. Alec has always stressed, and demonstrated by example, the importance of doing science that takes us in new directions, and he recognized early on and demonstrated repeatedly the enormous value of experiment and theory working together. He has been critical - and rightfully so - about the trend of "chasing the numbers", as this encourages scientists to mainly work in popular fields, where many others are active already, and he has mused about the citation impact of a scientist who is swimming against the stream. Nevertheless, let us mention one number: Alec has coauthored about 300 scientific papers that have appeared in the most respected journals in our field as well as in the highest-profile interdisciplinary journals. Apart from his papers, he is well-known for his lucid scientific lectures, typically showing novel, high-quality experimental results that define the state-of-the-art in the field and that challenge the current theory. What sets him apart from many other experimentalists is the impressive set of diverse topics that he has covered over the years, some of which we will mention below.

As a PhD student of Yuan T. Lee at UC Berkeley, working alongside Dan Neumark, Alec was involved in what have become textbook molecular beam studies of the F + H_2 reaction, in which the first indications of reaction resonances were obtained. It is difficult to imagine a better intellectual environment and a more promising and exciting start of someone's scientific career than Alec's. The photograph of Yuan Lee standing next to his molecular beam scattering machine with Alec and Dan sitting on top of it, taken about 40 years ago, has become an icon in the international molecular beams community. Alec received his PhD from Berkeley in 1986, the year that Yuan Lee was awarded the Nobel Prize in Chemistry.

As a postdoctoral scientist in the group of Peter Andresen within the department of Hans Pauly at the Max Planck Institute for Fluid Dynamics in Göttingen, Alec explored the use of tunable ArF and KrF excimer lasers in gas phase molecular physics experiments. Nowadays, these lasers are hardly available anymore, but, by producing several hundred mJ of energy in an ∼10 ns duration pulse with a spectral bandwidth of about 0.5 cm^{-1}, these lasers offered truly unique possibilities, in spite of their limited tuning range of only about 1 nm. Alec showed that, for instance, molecular oxygen, nitric oxide, carbon monoxide, and the hydroxyl radical can all be resonantly excited within the approximately 200 cm^{-1} wide tuning ranges of these lasers around 193 and 248 nm by driving dipole-allowed transitions

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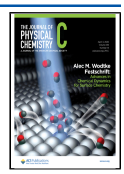

with weak Franck−Condon factors or spin-forbidden transitions.

Alec realized that these tunable excimer lasers would be ideally suited as the pump laser in various stimulated emission pumping (SEP) schemes, thereby enabling the efficient production of molecules in highly vibrationally excited levels. From his start-up funds at the University of California at Santa Barbara, he purchased a tunable excimer laser, and he performed a series of ground-breaking experiments using highly vibrationally excited molecules as study objects and as scattering partners. In studies on the state-to-state collisional energy transfer in highly excited NO and O_2 molecules, multiquantum vibrational relaxation was found to be important, and anomalously high self-relaxation rate constants were observed. He discovered that highly vibrationally excited O_2 molecules ($\nu \geq 26$) can react with ground-state oxygen molecules to form ozone. When such highly vibrationally excited oxygen molecules are produced in the photodissociation of ozone, their reaction with ground-state oxygen molecules amounts to an autocatalytic process for enhanced ozone production. The neglect of this process can explain why the stratospheric ozone concentration in earlier models is less than what has actually been observed (the "ozone deficit" problem). In another seminal experiment, aimed at spectroscopically investigating an isomerization reaction, Alec used the ArF laser in combination with a tunable dye laser to populate via SEP a large number of vibrational levels in hydrogen cyanide. Levels containing up to 2.3 eV of vibrational energy in the electronic ground state could be populated, and at the highest energy the signatures of delocalized, HCN-CNH isomerizing vibrational states were identified.

Although not mentioned in his autobiography, Alec spent a few months of his sabbatical in 1993 at the University of Nijmegen, to work with one of us (GM). We had no clear plan which experiments to perform during his sabbatical, but when Alec arrived in The Netherlands, he was excited about a presentation he had just heard from Richard Saykally on cavity ring-down spectroscopy. Over a few drinks in a local bar, the plan was hatched to use a variant of this method to sensitively detect the stimulated emission transition in a SEP setup, i.e., to try to perform cavity ring-up spectroscopy. The experiments on CS_2 that were then set up in the weeks after that showed that it is difficult to preclude lasing when SEP is performed in a high-Q optical cavity, something that, in hindsight, should not have come as a surprise. In the end, this work resulted in a paper on cavity ring-down spectroscopy in relatively short, stable optical cavities with an almost continuous mode spectrum, an approach that has since then been followed by various research groups for sensitive absorption measurements.

As an example of yet another class of experiments that Alec performed, we'd like to mention the photochemical production of cyclic N_3 from methyl azide, which was featured on the cover of this journal in February 2008. The cyclic structure of N_3 was already postulated by Theodor Curtius in 1890 but was only found by Alec's group, in their velocity map imaging experiment. Both of these brief episodes are exemplary of Alec's career, in the course of which he entered a variety of new fields and tackled diverse problems out of pure curiosity, designed and set up original experiments, and has made highly recognized and lasting contributions.

Highly vibrationally excited molecules have been a common thread in Alec's research. In a series of ground-breaking experiments, Alec and his co-workers studied the influence of molecular vibrations in promoting electron transfer reactions at a metal surface. When colliding highly vibrationally excited NO molecules with a metal surface, they observed highly efficient multiquantum vibrational relaxation of NO, and they reported the efficient ejection of electrons from the metal surface; they even measured their energy distributions. These experiments demonstrated unambiguously the direct conversion of vibrational-to-electronic excitation, and so became a clear manifestation of the breakdown of the Born−Oppenheimer approximation - one of the basic assumptions used in theoretical descriptions of bond-dissociation at metal surfaces.

Since his appointment as director of the Max Planck Institute for Biophysical Chemistry (now renamed into Max Planck Institute for Multidisciplinary Sciences) and as Professor at the Institute for Physical Chemistry at the Georg August University in Göttingen in 2010, Alec has further expanded his activities, with a focus on the kinetics of reactions on surfaces. In his "Dynamics of Surfaces" department, a variety of experiments have been set up in which quantum-state, angle- and speed-resolved beams of atoms and molecules interact with surfaces. Catching molecules in the act of reacting and comparing this with the outcome of first-principles theoretical simulations yield "the world's greatest microscope", as Alec put it when he received the Gerhard Ertl Lecture Award in Berlin in 2022 as well as when he gave the Yuan T. Lee Lecture in Physical Chemistry in Berkeley in 2023.

Many more spectacular results are expected to emerge from his laboratory in the coming years, not at least because he recently secured a large ERC Synergy Grant (together with Peter Saalfrank, Liv Hornekær, and Varun Verma) to investigate how molecules observed by space telescopes are produced by chemical reactions in and on interstellar icy dust grains. Infrared spectra of molecules in these ice grains will be measured, and their chemical reactivity will be studied under interstellar-space conditions. The use of superconducting nanowire single-photon detectors that are crucial for this undertaking has been pioneered by Alec in recent years in a series of beautiful experiments on the vibrational energy transfer between CO molecules on a NaCl(100) surface. Using time- and frequency-resolved infrared fluorescence spectroscopy, vibrational energy pooling and orientational isomerization have been observed in this apparently simple but unexpectedly rich system.

It was an inspirational experience to work in Alec's group when he started at Santa Barbara for one of us (XY). Developing innovative instruments and experiments in efforts to investigate the most interesting scientific problems was a central theme in Alec's research career from the beginning. This approach has been highly effective in his lab to attack frontier scientific problems and was also indeed educational for his students and postdocs. Alec was always very excited about developing and applying new techniques to solve challenging scientific problems. For example, when the VUV FEL at Dalian (Dalian Coherent Light Source) was developed, he started immediately thinking about how this VUV FEL could be applied to study surface scattering dynamics. His novel idea led to a successful scientific collaboration project between the Max Planck Gesellschaft (MPG) and Chinese Academy of Sciences (CAS), which survived through the difficult COVID-19 pandemic period, thanks to his persistence. Alec's powerful and unwavering pursuit of international collaborations in basic scientific research between the east and the west has been truly admirable, especially in this current problematic world.

Alec is always challenging theoreticians to provide coherent and insightful interpretations of his experiments. Over the years,

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he has attracted around him a large theoretical community, who eagerly offer models and calculations to better understand the physics manifested in his beautiful experiments. He has never been a passive observer in these efforts but an active participant. His keen interest and deep insight have greatly contributed to the physical picture emerging from such joint experimental-theoretical studies, which have become a hallmark of his scientific work.

Many of the (under-)graduate students, postdocs, and colleagues who have worked closely together with Alec over the years have been eager to contribute to this *Festschrift* with their original publications in honor and appreciation of Alec's work and personality. Alec has been both a role model and an inspiring advisor for many of his students and postdocs, and he has made lasting impacts on their professional careers. Alec and his wife, Liesel, supported by their two sons, have always made a great effort to organize lively social events at their home, thereby bringing scientists from various nationalities and different communities together. At their home in Santa Barbara, one could end up in their swimming pool; at their home in Göttingen, one can enjoy their yard. The actual occasion for these parties can differ, e.g., Thanksgiving dinner, a round birthday (almost round would also qualify), soccer championships, celebration of a research grant or of an important publication, etc.; being your guests has always been equally delightful and entertaining. Alec, we thank you a lot for all you have done for the scientific community, and we hope that you will enjoy browsing through this issue and reading the various contributions by your friends just as much!

Hua Guo orcid.org/0000-0001-9901-053X
Gerard Meijer orcid.org/0000-0001-9669-8340
Xueming Yang orcid.org/0000-0001-6684-9187

AUTHOR INFORMATION

Complete contact information is available at:
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Notes

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