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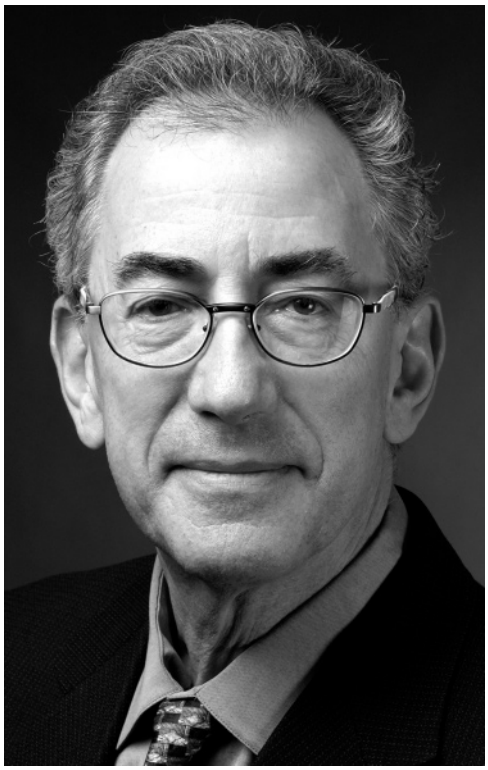

Tribute to Michael L. Klein: Scientist, Teacher, and Mentor

Michael Klein's field of research is computer simulation of condensed molecular systems. The 40 plus years of his scientific life span the birth and coming of age of this new discipline in science. In 2005, Mike marked his 65th birthday, and we are delighted to use this occasion to dedicate a special issue of the *Journal of Physical Chemistry B* to Mike. This issue celebrates the numerous important and remarkable contributions Mike has made to chemistry, biophysics, materials science, and, in particular, the field of computer simulation. It is also an opportunity to express our gratitude and to thank Mike for his tireless efforts on behalf of his students, postdocs, and colleagues in the international simulation community. Molecular simulation began with pioneering studies of simple monatomic fluids using first hard and then soft potentials. Next came molecular liquids such as water. The systems increased rapidly in size and complexity (e.g., type of interactions, conformational flexibility, and inclusion of quantum effects), eventually leading to molecular simulations of biological systems. Mike's scientific interests and activities mirror this evolution. The focus of his work has always been on "realistic models". He has proven himself a true master at using simulation to make models come to life, produce results that can be compared directly to experiment, and help us to understand what is going on in the "real world" (one of his favorite expressions). This vision of the role of simulation has guided and motivated Mike in his research. His keen scientific mind was quick to see the potential of new theoretical and technical developments. If not making the first step in the development of a new method, then he often made the crucial second step of applying it to the "right" system, showing what the method could contribute to solving problems of interest to theorists and experimentalists alike. In fact, few of the researchers in the computer simulation world have taken the interaction with experiment as seriously as Mike. Many of his projects have been directly inspired

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by discussions with experimentalists. While the confrontation with the reality of experiment is probably dearest to his heart, he also has an intense interest in method development, in particular those methods that enable him to approach experimental conditions more closely. As a result, his research group has produced over the years a number of remarkably original methods.

Though a chemist by education Mike's early research was firmly rooted in theoretical solid state physics. While a Ph.D. student in Bristol with Hugh Barron and later as a postdoc with George Horton at Rutgers, he studied the anharmonic dynamics of rare gas solids using some of the most formal and abstract instruments of theoretical physics (diagrammatic perturbation techniques). The demanding work on anharmonic lattice dynamics must have been a good preparation to appreciate the revolution brought about by molecular dynamics simulation. Mike was introduced to the method during his extensive visits to France working with Jean-Pierre Hansen, Dominique Levesque, and Jean-Jacques Weiss on solid argon and nitrogen. This was in the early 1970s, when Mike had already moved to Ottawa to become a member of the staff at the prestigious National Research Council of Canada (NRCC). A decisive moment in his conversion to molecular simulation, close to a revelation, was the Monte Carlo study by Barker and Watts on liquid water followed by Aneesur Rahman's seminal molecular dynamics simulation of liquid water. This was proof for Mike that computer simulation could access completely new territory, in particular, make the giant transition to "real" finite temperature disordered molecular systems, forging links between theory and experiment where this had not been possible before (and opening up the way for him back to chemistry). An important hub for the excitement and activity in the emerging computer simulation community was the Centre Européen de Calcul Atomique et Moléculaire (CECAM) established by Carl Moser in Paris. There, Mike found a place in a circle of enthusiastic people who were to become, like him, leaders in computer simulation (among them Berendsen, Ciccotti, Frenkel, McDonald, Ryckaert, and also Rahman himself). They met regularly to discuss the latest progress and problems, but there was also the hard work of coding and running of jobs (in those legendary days workshops could last for over a month).

Having established his own research group at the NRCC in Ottawa and urged on by his British friend and long-time collaborator Ian McDonald, Mike changed focus to condensed molecular systems. The goal was to understand the interplay between translational and orientational order/disorder in both molecular crystals and liquids. While molecular fluids of ever increasing complexity were to become the major theme of his research, Mike has never lost interest in solids. In fact, one of the highlights of his period at the NRCC was work with Shuichi Nosé on the high-pressure phase behavior of molecular solids, which is also a good illustration of Mike's approach to simulation. Realizing the enormous potential of the extension of Andersen's constant pressure method to solids by Parrinello and Rahman, he asked Nosé to adapt the method so it would be possible to treat interaction models of rigid molecules. After an investigation of the high-pressure phase transformations of nitrogen, he, in collaboration with John Tse, went straight on with a most challenging application, namely, the complicated behavior of ice under pressure (a material of special practical importance in Canada).

In addition to isobaric isothermal MD, Mike experimented with numerous other advanced simulation methods, often taking them almost literally out of the hands of their inventors and preparing the new tool to attack new problems. In this context we can mention that Mike was the first to exploit the path integral molecular dynamics method for the study of solvated quantum particles in polar solvents (after again Parrinello and Rahman showed the way with a study of an excess electron in a molten salt). This project led to a collaboration with David Chandler and encouraged him to consider joining the highly active theoretical chemistry community south of the border, which he eventually did.

Another exploration with Ottawa origins, which turned out most fruitful for his future research, was the collaboration with Jean-Paul Ryckaert on flexible long-chain molecules (alkanes). Mike was keenly aware that in order to get this right, the crucial test was not the liquid, but the delicate structure of the solid (orientation, layering, tilts, etc.). He started a careful and exhaustive series of validations of model potentials. In the back of his mind was probably already the study of aggregation of amphiphiles in solution, combining the modeling of long-chain molecules with the modeling of the interactions in aqueous solution. This massive project dominated the first few years of Mike's research endeavors after moving to the University of Pennsylvania in 1987. This move marked the beginning of a period of accelerated expansion of the scope of Mike's research. His group at Penn seemed to be interested in, and also successfully mastered, almost everything that was at the frontier of computer simulation. The work on amphiphiles evolved in a beautiful and spectacular series of simulation studies on micelles and self-assembled monolayers. Model systems grew bigger all the time, soon reaching the size of full membranes including membrane proteins (in particular ion channels), which even attracted the attention of colleagues in the medical school at Penn. However, Mike also returned to his solid state and materials roots in a project on the orientational order/disorder transition in fullerenes. This investigation was carried out in close contact with experimentalists at the Laboratory for the Research on the Structure of Matter (LRSM). The affiliation with the LRSM represented a huge temptation. It appealed to Mike's strong affinity to experiment and also his talents as a leader. This went as far as accepting, in 1993, the directorship of the LRSM. This demanding position required him to organize the research proposals of an entire lab of experimentalists and a few theoreticians in a package that would maximize their impact. He is still, after more than 12 years on duty as director of the LRSM, not ready to resign (as far as we know).

The directorship of the LRSM was not allowed to get in the way of broadening his research program in computational chemistry at Penn's Department of Chemistry. Car−Parrinello molecular dynamics simulations of chemical reactions could certainly not be missing in Mike's vision of an all-round modeling group, and he extended his research also in this direction. The Car−Parrinello activities at Penn initially focused on benchmark studies of small vacuum systems and the acid−base chemistry (proton transfers) of aqueous solutions and other hydrogen bonded solvents. Part of this work was carried out in collaboration with Michele Parrinello's group, first in Zurich and later in Stuttgart. This led to a very productive exchange of people and ideas and also gave Mike the chance to occasionally escape his duties at Penn for another visit to Europe. However, after the first encouraging experience with biosystems in his classical modeling projects, Mike became fascinated by the wide range of problems and opportunities this field offers to computer simulators and steered the Car−Parrinello efforts in his group in the direction of bioapplications, in particular the study of enzyme catalysis and, again, ion

channels. Mike resolved the obstacle of the rather steep learning curve for using this technically very complicated method in a characteristic way, namely, by a clever exchange of talented young people with other research groups, which now also included the groups of former students and postdocs.

All this activity became bundled in the Penn Center for Molecular Modeling (CMM) which Mike founded in 1995. This center has become a beehive for simulation and modeling. The CMM has taken a special interest in the development and application of coarse grained models for very large scale biosystems. The work on membranes convinced Mike that atomistic modeling has its limits after all and that tackling the kind of problems he encountered in his discussion with experimentalists required a multiscale approach. The Klein group happened to be in a most favorable position to embark on such an ambitious development. The experience and vast database of results of atomistic simulations on amphiphilic systems formed a perfect point of departure and a testing ground for the parametrization of coarse grained potentials. This is not an easy task, requiring keen insight in what matters and what can be ignored. This was therefore a project ideally suited to Mike's unique sense for modeling condensed molecular systems. The first results produced by the coarse graining project confirm that simulation has indeed entered a new regime of phenomena.

This is an appropriate place to add a further comment on Mike's attitude with regard to method and code development. Characterized by a curious modesty, sometimes bordering on detachment, this attitude gives the advantage of maximum flexibility. He uses a whole range of different tools, some developed in his own group, some taken from elsewhere. At the same time, he leaves the young people in his group all freedom to pursue their ideas if they want to, encouraging them to do so and making occasional crucial suggestions. He keeps, however, a certain distance and makes no claims for himself. This approach has a wonderfully stimulating effect on creative and scientifically independent young people for whom Mike's group holds a special attraction, producing a number of major innovations, including thermostats and simpletic molecular dynamics integrators (for equilibrium and nonequilibrium systems).

This brings us to the exceptional role Mike has played as teacher and mentor. A large number of beginning computational scientists (chemists, physicists, and applied mathematicians) from all over the world have passed through his group as students, postdocs, or visitors. They were given the opportunity to participate in first-rate scientific research and, in the process, learn a few vital techniques of the trade of computer simulation, from Mike himself and through interactions with other group members. The experience persuaded a large fraction of the young people under his wing to continue on in science and computer simulation. Here you could count on Mike too. You were assured of his professional advice and support during the stay in his group, and even more important, after leaving. Many of these people have become successful independent scientists, established their own research groups, and have even started sending a second generation of students and postdocs over to be educated by Mike and, in turn, prepared for the "real world". Because of his close ties with Europe, he is in many ways straddling the Atlantic, Mike's mentorship was especially effective for his European pupils. The European connection also showed in his interest in and concerns for the well-being of research groups and institutions in Europe (and we could also perhaps include here Canada). The European computer simulation community is particularly grateful for his contributions to the success (and survival) of CECAM. In his many advisory functions, Mike showed great wisdom and tact, which of course meant that he was asked again and again.

Finally, we would like to thank Mike and Brenda for the welcome and generosity they have shown to all members of his large group. The summer parties at the Jersey shore are memorable occasions.

On behalf of all his students, postdocs, and colleagues, we wish Mike continued success in, and enjoyment of, his research.

Michiel Sprik, Ilja Siepmann, Doug Tobias, and Mark Tuckerman

Guest Editors