

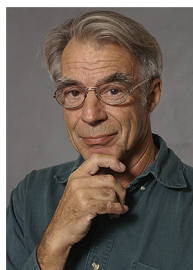


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Charles Austen Angell, 1933–2021



One of the giants of physical chemistry, C. Austen Angell, passed away on March 12th, 2021 in Tempe Arizona. He was internationally recognized as a luminary in the fields of glasses, water and ionic liquids. Over a career lasting more than 60 years, Austen's seminal contributions refashioned these disciplines many times over and produced lasting impacts that will shape the development of these areas for years to come. Fueled by his broad scientific curiosity and his firmness of purpose, Austen traversed multiple fields of science and brought innovative viewpoints to solve both fundamental and applied problems.

Austen's passion and enthusiasm for science was unparalleled. Throughout his remarkable career, he inspired multiple generations of students and colleagues to explore new problems with an open mind. He is widely remembered for his singular ability to engagingly listen to others. His genuine interest and intuitive feel for the fundamental significance of a colleague's work made him an incomparable collaborator. He was the personification of what is best in a scientist, combining passion, humility and integrity with deep scientific knowledge and insight. His genius consisted of perceiving patterns in the fabric of knowledge that had gone undetected, but which seemed so compelling, once he pointed them out, that they could not be but embraced by the community. This made him a role model for so many young scientists across the world.

His ability to observe and to transcend ingrained boundaries between diverse fields of study and recognize the patterns they have in common led him to make critically important contributions across the chemistry, physics, and materials science communities. His aptitude for explaining deep concepts in his talks and papers earned him well-deserved recognition from colleagues worldwide. Although he was widely regarded as a visionary, Austen always remained remarkably humble. The strong/fragile classification of liquids introduced by Austen is now a staple of the glass and liquid state sciences and is used in industry to help improve manufacturing (e.g. from vaccine vials to 3D printed amorphous metals) and predict relaxation processes, as well as in academic work to help frame the principles of structure-property relationships in liquids and glasses. Yet, Austen often wondered if his

idea “may have got more attention than it merited.” His humility was as legendary as his talent.

The “Angell plot” of glass-forming liquids is probably Austen's most celebrated emblem. It provided a conceptual framework for classifying all categories of liquids in one universal diagram in terms of the “fragility” and as such created a new paradigm for glass science that clarified and unified many disparate schools of thought, and attracted a broad community. In particular, his views of the glass transition from the perspective of liquid dynamics had a deep influence on the statistical mechanics and physics community. He later advanced the concept one step further by demonstrating that some key physical systems such as water actually underwent a fragile to strong transition upon cooling. This in turn, brought to light the concept of polyamorphism, which remains a deeply enigmatic challenge in categorizing the states of matter.

Austen's pioneering studies of supercooled and stretched water launched the modern era of investigation into the anomalies of water, and introduced ideas that continue to influence new thinking in this area. The importance of his work on water notwithstanding, the great height from which Austen viewed the scientific landscape allowed him to seamlessly move on to consider a much wider spectrum of liquid-state behavior, encompassing other network-formers such as silica and silicates, and placing water in context along this spectrum. For instance, water-like liquid anomalies have recently been recognized in technologically relevant phase-change materials and have been linked to the characteristic switching cycles of non-volatile phase-change memory devices based on these materials.

Austen also produced seminal studies of highly decoupled motions of small weakly charged anions and cations in the now burgeoning field of ion conducting solids, especially glasses. He was the first to recognize that the short time extrapolation of the dynamics of mobile ions led inextricably back to the quantum controlled harmonic vibrations of these ions. This insight into and connection between the long time d.c.-limit of the conductivity and its quantum controlled vibrational limit enabled Austen to predict and understand for the first time the ultimate high conductivity limit of such solid electrolytes. This upper limit he predicted in the mid-1980s remains today as a singular challenge to battery technologists worldwide, as they seek safer yet more energy-dense and more rapidly charged all-solid-state batteries that will revolutionize portable electronics and enable electrified carbon-neutral transportation.

Austen was a progressive thinker, aware of the accelerating pace with which new research methodologies became available. He interpreted any glimpse of uncharted territory as an invitation to explore. Although by trade an experimental chemist, he was one of the first to embrace molecular simulations as a means to gain new insights into the amorphous state of matter and the phase behavior of liquids. Prior to this, structural models of glassy materials were constructed by hand and

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analyzed using a yardstick, consuming the time allotted for a PhD thesis project for a mere handful of systems. Conversely, the computational approach to realizing such models accelerated the pace of gaining the desired insights enormously, which had a major influence on theories of the glass transition and structure-property relationships in amorphous materials.

Austen was always a friendly, approachable and memorable presence at conferences. He made a special effort to connect with young researchers, an experience that many remember fondly, and which could change the course of a career. Ever-present in the front row, Austen would have a question after every talk. With a characteristic twinkle in his eye, he would raise his hand, index finger extended. Simultaneously, he would always look back toward the audience, to make sure he wasn't preempting a question from another. This simple gesture simultaneously captured his delight for science and his respect for his community.

Studying under Austen's tutelage or engaging with him in a scientific collaboration was not just a joint effort in the quest for discovery. It revealed a new dimension of intellectual pursuit, opening a vast space of opportunity, while at the same time fueling one's exploratory spirit through Austen's trust and support. He is remembered by his students and postdocs as a thoughtful and caring mentor and they will forever cherish the lasting presence of his wisdom. He always encouraged his students and postdocs in independent thinking and exploring new scientific territories, and then cheered their achievements and promoted their careers. Typical of Austen's PhD mentoring, many of us will forever remember and be impacted by Austen's ever-open door to invited guests and scientists from around the world. Such visits often resulted in reciprocal visits by his PhD students to his visitors' laboratories and universities. While at the time, these experiences seemed like an exciting added bonus and a welcome distraction from the academic routine, the true value became apparent only with time, for it meant the beginning of belonging to a supportive network of like-minded individuals who all

give continuity to Austen's aspirations. From Purdue to Arizona State University, Austen trained generations of scientists, many of them becoming professors like him, who in turn trained future professors. At the time of his passing, Austen had several great-great-grand PhD students. His legacy is already enormous, and is bound to grow in the future.

Beyond his brilliance as a scientist, Austen was also a champion at life itself. From driving across the deserts of Africa as a young man to hiking mountains all over the world with friends and family, Austen never missed an opportunity to seize the day. Most remarkably, his spirit never aged. He will be sorely missed and dearly remembered.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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