

State-Specific Chemistry

Photodissociation Dynamics. Spectroscopy and Fragmentation of Small Polyatomic Molecules. REINHARD SCHINKE. Cambridge University Press, New York, 1993. xvi, 417 pp., illus. \$89.95 or £50. Cambridge Monographs on Atomic, Molecular, and Chemical Physics.

In the closing paragraphs of his famous textbook *The Principles of Quantum Mechanics* Paul Dirac wrote: "The domain of applicability of the [quantum] theory is mainly in the treatment of electrons and other charged particles interacting with the electromagnetic field—a domain which includes most of low-energy physics and chemistry." This kind of stuff does little to reduce the sense of unease that plagues some chemists who believe their discipline is, in principle, ultimately reducible to quantum physics. It feeds the prejudice that chemistry is somehow second-class, inferior to the first-class fundamentalism of physics.

This perception of chemists scrambling for the leavings of theoretical physicists is perhaps nowhere stronger than in the area of chemical reaction dynamics. The elementary act of chemical transformation lies at the center of the chemists' domain. Many theories of chemical change—often little more than pictorial representations making use of dots and arrows to show the "movement" of electrons—appear hopelessly naïve compared to the elegant mathematics of the quantum formalism. The notion that the "true" description lies at the quantum level is compelling: nobody would deny that the heart of the act of chemical change beats with a quantum beat.

Chemists salvage some self-respect by pointing to the difficulty—if not the sheer impossibility—of applying quantum theory's complex mathematical structure to most systems of chemical interest. Principle is one thing, they say, practice another. But the reality is even more heartening. Over the last 10 to 20 years it has been the experimental chemists who have done the most to expose the quantum nature of chemical change. The theoreticians have been running hard to catch up.

The tremendous progress that has been made in the laboratory derives primarily from developments in technology (particularly laser technology) and careful selection and tailoring of research programs. Small molecules in the gas phase or in the ultra-cold environment of a supersonic expansion are amenable to ultrafast or high-resolution laser spectroscopy studies that provide information resolved at the atomic level of motion or at individual molecular quantum states. Photodissociating such molecules

through selected quantum states allows the chemist to probe the last half of the elementary act of chemical change—the "half-collision"—in unprecedented detail.

In *Photodissociation Dynamics* theoretician Reinhard Schinke provides the research scientist with the essential tools of this trade. For the budding theoretician, here are the various approaches to the theoretical description of the photodissociation of small molecules expertly developed from undergraduate quantum mechanics. For the budding experimentalist, here is the lingua franca—the common theoretical language—that has long dominated this research field and provides the accepted means of interpreting and communicating the results of experiments.

The book has three main components. A broad general overview of the basic theory is followed by a series of detailed discussions of its application in the calculation of such observables as absorption spectra (structured and diffuse), photodissociation cross sections, and photofragment state and energy distributions. In a friendly but authoritative way, Schinke illustrates the different perspectives available from the time-dependent and time-independent quantum treatments and discusses their relative merits. In the closing chapters he examines the photodissociation of van der Waals molecules and vibrationally highly excited molecules, emission from dissociating molecules, nonadiabatic transitions, and the relatively new experimental science of femtosecond time-resolved chemical dynamics. Throughout the book he compares and contrasts the theory with examples from the "real" world.

If I have one criticism, it is that Schinke seems to portray a science in maturity. The reader receives the impression that experiment and theory are closely in line, the few differences here and there fixable through some tweaking of the relevant *ab initio* molecular potential energy surfaces. It would have been useful to point out a few areas of future challenge, areas in which the ambitious researcher can seek to make a mark. Experiments in real time are now revealing molecular quantum behavior in impressive detail, bringing the transition state—once no more than a theoretical abstraction—directly into the world of experience. At the same time, the practical dynamicist must become increasingly suspicious of the many approximations necessary to bend the quantum formalism to the task of prediction, beginning with the assumption of quantum molecules in a classical radiation field. And how long will it be before some of the more well-known conceptual problems and paradoxes of quantum theory become manifest on a molecular scale?

But these are quibbles. Schinke has dis-

tilled a vast and complex scientific literature into an accessible monograph. This book is destined to become standard reading for graduate students about to embark on research careers in chemical dynamics, as well as reflective reading for more mature scientists who thought they understood their subject.

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