Design Challenges for Control of Molecular Dynamics

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Molecular Surgery and Synthesis

Since the advent of lasers the notion of actively controlling dynamics at the molecular scale has become a subject of intense activity.Material modification at the molecular scale is a problem of control over molecular structure, if not dynamics itself. The successful introduction of modern control techniques at the level of quantum systems should open up a new domain of technological advances. One possible application is in the storage and retreival of information through tailored molecular scale transformations. Control design, combined with the appropriate surgical tools from laser physics and quantum electronics, could have a direct impact on many electronic materials by providing means for probing, altering, and eventually synthesizing new products at the molecular scale.

The original logic put forth in the early 1960s for using lasers to create a molecular "pair of scissors" can be succinctly stated as follows: a) determine the spectrum (particularly vibrational) of a molecule of interest; b) interpret the spectrum in terms of local mode frequencies and identify the bonds of interest; c) find a laser to pump intensely at the required frequency and wait for the bond to break or an attacking reagent to preferentially react at the activated site. A number of clever variations on this theme have been considered but the essence has always been the same. Generally two results were found: *either* rapid redistribution of energy throughout the molecule prevented preferential chemical reactivity from focusing at the desired site in the molecule, *or* so much energy was deposited that the molecule literally exploded into fragments.

Therefore, Laboratory studies only served to prove that if answers (i.e., optical fields capable of achieving molecular scale objectives) actually exist, they cannot be determined based on intuitive arguments alone. In particular, molecules are capable of undergoing very complex dynamical motion and if this motion is to be controlled, the external optical field must literally take over the molecular dynamics, or at least work cooperatively with the dynamical capabilities of the molecule. Hence, one can see that any solutions to these problems, except for the most trivial cases, will inevitably call for fields having very complex temporal and/or frequency structure.

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Optimal Control — A Theoretical Framework for Exploiting Molecular Dynamics

Recent research [1]-[3] into optimal control of molecular motion attempts to put the subject on a sound theoretical footing and open the topic for careful systematic study. The optimal control framework provides a rigorous basis for assessing the practicality of achieving a given molecular objective and the means to design optical fields in favorable cases.

Having recognized that the tools of systems analysis [1],[3]-[6], and particularly control theory, provide the proper mathematical framework for translating physical objectives, constraints, and costs, there has recently been intense investigation of a number of physical problems [2],[7]-[9]. These examples illustrated the power of control theory in achieving molecular objectives, such as dissociation. In [1],[3]-[5] issues of controllability, existence of optimal fields, numerical approximations, and implementation are analyzed. In [6] a survey of the techniques of systems science as they relate to problems in molecular dynamics and control is given. At this early stage of development examples involving electronic, vibrational and rotational motion have been treated. In addition, the issues of well-posedness and robustness have been considered [1],[3]. Preliminary efforts in the last few years has clearly established the promise of this approach.

An important issue to understand in the present context of molecular control is the distinction between *ad hoc* intuitively designed schemes of control and those of optimal control. Intuitively designed molecular control schemes have been proposed typically based on the assumptions of perturbation theory and simple interference processes [10], and verification of the role of interference processes has been observed in the laboratory [11]. Although the advances in molecular optimal control also critically play on the essential structure of quantum mechanics with its wave nature giving rise to constructive and destructive interferences, it is, however, only through the more flexible approach of optimal control theory that the multitude of competing physical and chemical demands as well as laboratory constraints can be disentangled to seek viable solutions. Furthermore, realistic problems of control at the molecular scale are not often expressed solely in terms of an objective at a particular bond. Rather, a host of other auxiliary criteria can arise including the desire for minimal disturbance or damage to the remainder of the molecule, minimal expenditure of laser energy, the imposition of laboratory constraints, etc. Thus, one can see that the establishment of successful optical fields for controlling motion at the molecular scale is a complex balancing act requiring the careful use of modern control and design techniques.

For engineers who use control design techniques to solve complex problems ranging from aircraft design to robotic system control, the use of optimal control in such a context would seem to be routine . However, the unique structure of molecular quantum dynamical systems provides new challenges to the traditional paradigm of control design. For example, typically interesting dynamical events will occur on the ultrafast time scales 10^{-9} to 10^{-15} s, eliminating the possibility of simultaneously monitoring the evolution of the molecular state and feeding back that information to redirect the driving field. Since it will not be possible to have observation of the system on a real time scale, open loop controllers will be needed and this places an enormous demand on the correctness of the theoretically designed fields for laboratory implementation. This issue of robustness must

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be addressed properly if laboratory success is to be achieved.

New Probe Tools

The objective of designing optical fields is essentially to produce a molecular pair of scissors, and thus it is important to understand this subject in a broader context. Although ultimately demonstrating in the laboratory the ability to selectively break bonds by carefully designed optical sources would certainly be a notable achievement, its practical significance on an industrial synthesis scale may still be lacking due to the expense of executing the effort. Furthermore, the serious need for design robustness, mentioned above, raises the question of whether we currently understand Hamiltonians and optical coupling coefficients well enough to reliably design fields that can produce the desired objectives. However, the same tools of molecular control may possibly be turned around to design experiments for filling in this missing information where necessary. In particular, the optimal control concepts could ultimately be applied to design specialized pump-probe spectroscopy measurements for the purpose of inversion to yield missing molecular scale Hamiltonian information.

Although it is premature to specify detailed particular applications of successful molecular control, there may be many practical and fundamental consequences from understanding and establishing control at the molecular scale. Just as control tools are now being extended to the large scale realm of massive space station structures, the control community has much to offer and learn from efforts focused at the other extreme of the microworld. Indeed, one can argue that establishing limits of control at the molecular scale is a subject of basic interest which must be pursued for fundamental reasons alone.

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Conference Calendar

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Rensselaer's Third International Conference on Computer Integrated Manufacturing, May 20-22, 1992, Rensselaer Polytechnic Institute. Contact: Prof. A. Desrochers, Program Chair, Rensselaer Polytechnic Institute, CII 8015, Electrical, Computer, and Systems Eng., Troy, NY 12180-3590. Phone: (518) 276-6718.

IFAC Symposium on Intelligent Components and Instruments for Control Applications SICICA '92, May 20-22, 1992, Málaga, Spain. Contact: Prof. A. Ollero, SICICA '92, Facultad de Informática, Plaza El Ejido s/n 29013 Malaga, Spain, (34)-52-131412, FAX (34)-52-264270.

INCOM '92 - 7th IFAC/IFIP/IFORS/-IMACS/ISPE Symposium on Information Control Problems in Manufacturing Technology, May 25-28, 1992, Toronto, Canada. Contact: Mrs. Nicole Léger, INCOM '92, National Research Council Canada, Montreal Road, Building M-19, Ottawa, Ontario, Canada K1A 0R6, Telephone: (613) 993-9431, FAX: (613) 957-9828, Telex: 053-3145.

IEEE International Symposium on Industrial Electronics, 25-29 May, 1992, Xian, China. Contact: Professor J.D. Irwin, Department of Electrical Engineering, Auburn University, Auburn, AL 36849, U.S.A. Tel. 1(205) 844-1810, Fax. 1(205) 844-1809; or contact: Dr. Allen C. Chens, A.T.&T. Bell Labs, Rm. 1E134A, Whippany Road, P.O. Box 903, Whippany, NJ 07981-0903.

IFAC Workshop Automatic Control for Quality and Productivity, June 3-5, 1992, Istanbul, Turkey. Contact: Prof. Talha Dinibutun, Dean, Mech. Engrg. Dept. Istanbul Technical University, Gumussuyu 90191, Istanbul, Turkey. Tel: 90-1-1456073; Fax: 90-1-1450795.

International Joint Conference on Neural Networks (IJCNN), Baltimore, MD, June 7-11, 1992. For further information contact: IJCNN'92, Meeting Management, 5665 Oberlin Drive, Suite 110, San Diego, CA 92121 U.S.A. Telephone: (619) 453-6222. FAX: (619) 535-3880.

IFAC/IFIP/IFORS/IEA Symposium on Man-Machine Systems, June 9-11, 1992, The Hague Netherlands 1990. Prof. P.L. Brinkman Delft Univ. of Technology Fac. of Mech. Engg & Marine Tech. Mekelweg 2, NL-2628 CD Delft, NL.

IFAC Workshop on Artificial Intelligence in Real-Time Control, June 16-18, 1992, Noordwijkerhout, Netherlands. Prof. ir. H.B. Verbruggen Delft Univ. of Technology fac. of Electrotechn., POB 5031 NLO-2600 GA Delft, Netherlands.

IFAC Workshop Real-Time Programming, June 23-25, 1992, Bruges, Belgium. Prof. L Boullart, State Univ. of Ghent, Automatic Control Lab., Grot Steenweg Noord 2, B-9710 Gent-Zwijnaarde, Belgium.

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