CYBERNETICAL PHYSICS
AND CONTROL OF MOLECULAR SYSTEMS[1]

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1. INTRODUCTION. PHYSICS AND CYBERNETICS

Looking into the past. Encyclopedias define physics as the science studying the Nature, specifically its basic and most universal properties. The age of physics is about two millenia, and its history may be traced back to ancient times. Indeed, the term ``Physics'' meaning ``Nature'' in Greek was introduced by Aristotle.

Cybernetics is much younger and the date of its birth is known precisely. Though the term also was coined in the Ancient Greece, the foundation of cybernetics as a science is associated with publishing the seminal Norbert Wiener's book [1] in 1948. Wiener defined cybernetics as the science of control and communication in animal and machine. Novadays cybernetics is understood as control and steering theory in a broad sense, including different methods and approaches born within control theory during half of century of its extensive development, such as identification, estimation, filtering, information theory, optimization, pattern recognition, etc [2].

Both physics and cybernetics were emerging areas in the XX century and contributed a lot into modern science. However, cybernetical terms were rare guests at the pages of physical journals until recently; its influence on physical researches has been neglectible. The reason lies, perhaps, in totally different methodologies of the two sciences. Physics (e.g. mechanics) is a classical *descriptive* science, while cybernetics (e.g. control theory) represents a sort of paradigm for *prescriptive sciences* [3]. It means that the main aim of physics is to describe and analyze a natural system, while the aim of cybernetics is to transform a system by means of controlling action in order to achieve its prescribed behavior.

It is worth noticing that automatic and automated systems are used in physical experiments for a long time. Even more, no one serious physical

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experiment is performed without usage of automated equipment. However, control system usually plays a secondary role, just providing a prespecified mode of the experiment. No creative interaction of physics and control theory arose. No cybernetical methods were used to discover new physical effects and phenomena. Surprisingly, the situation has changed in the 1990s when two new areas emerged: *control of chaos* and *quantum control*.

Control of chaos. New avenue of research in physics was opened in the beginning of the 1990s by results in control and synchronization of chaos. It was discovered by E.Ott, C.Grebogi and J.Yorke [4] from the university of Maryland that even small feedback action can dramatically change behavior of a nonlinear system, e.g. turn chaotic motions into periodic ones and vice versa. The idea has become popular in the physics community almost instantaneously. Since 1990 hundreds and hundreds of papers were published demonstrating ability of small control (with or without feedback to change dynamics of real or model systems significantly, even qualitatively.

By the year 2003 the number of quotations of the paper [4] exceeded 1300 while the total number of the papers related to control of chaos exceeded 4000. The number of papers published in peer reviewed journals achieved 300-400 papers per year by the beginning of the XXIst century.

Interestingly enough that the general problem of suppressing chaos in a nonlinear system by means of a periodic controlling action was first posed in the papers [5], [6] published a few years earlier than [4]. A solution to the problem based on computer simulation was proposed in [5], [6] for a model ecological system of 4th order. Moreover, the possibility of transforming chaotic behavior of the Lorenz system into a periodic one under harmonic excitation was discovered still in 1983 [7]. However, the papers [5], [6], [7] did not trigger any stream of publications despite being translated and published in the West. The method proposed in [4] is now called OGY-method by its authors' initials.

It is important that the obtained results were interpreted as discovering new properties of physical systems. Thousands of papers had been published aimed at studying properties of systems by means of control, identification and other cybernetical means. It is important that overwhelming part of those papers are published in physical journals while most authors represent physical departments of the universities. The above facts provide evidences for existence of the new emerging field of research related both to physics and to control. In 1999 it was suggested to call it **Cybernetical Physics** [9], [10]. A concise survey of cybernetical physics is presented in [11].

However the potential of modern nonlinear control theory still was not seriously demanded although the key role of nonlinearity was definitely appre-

ciated. On the other hand, control community was not too fast in offering its potential to a new control application area. The reason is, perhaps in that new problems often differ from conventional engineering control problems. Indeed, instead of classical regulation problem (driving a controlled system trajectory to a desired point) or tracking problem (driving a controlled system trajectory to a desired motion) new types of control goals arise: creation of modes with partially specified properties (synchronization, transformation of a chaotic motion into a periodic one, etc.) On the other hand, more strict restrictions are imposed on controlling action which correspond to physically motivated requirement of minimum intervention in the natural evolution of the physical system ("small control" requirement).

Later it has become clear that such type of control goals are important not only for control of chaos, but also for control of a broader class of oscillatory processes. It lead to a development of a unified control framework for oscillatory (including chaotic) systems [8]. A natural next step is to pose a a general control problem of studying properties of physical systems that can be created of modified by means of (small) feedback actions [9], [10].

Recently an interest in application of cybernetical methods to search of new physical effects is observed in other fields of physics or mechanics: control of quantum systems, control of lasers, control of plasma, vibrations control, beam control, control in thermodynamics, etc. As a consequence, a number of control related papers in physical journals is growing rather rapidly. Especially fast growth of activity during last decade is observed in control of molecular and quantum systems.

Control of molecular and quantum systems. Perhaps, it is the area where ideas of control appeared first. One may trace its roots back to the Middle Ages, where alchemists were seeking for ways to change a natural course of chemical reactions in attempt to transform lead into gold.

The next milestone was set by the famous British physicist James Clerk Maxwell. In 1871 he introduced a hypothetical being with ability to measure velocities of gas molecules and to direct fast molecules to one part of the vessel, keeping slow molecules in the other part. It produces a temperature difference between the two parts of the vessel which seems to contradict to the 2nd law of thermodynamics. Now we know this being under the name ``Maxwell's Demon'' introduced by another famous physicist William Thomson (Lord Kelvin). The seeming breaking of the 2nd law helps to its better understanding. Now, after more than a century of fruitful life, Demon is even more active than in the past. In the XXth century Demon was exploited by L.Szillard, D.Gabor, L.Brilluin and others who studied the interplay between energy (entropy) and information [12], [14], [15], [16]. It has helped to the humanity to

realize that a measurement or computation requires some dissipation of energy. Attempts to diminish dissipation led to the idea of quantum computers [16], [17], [18], [19]. Recently the ways of experimental implementation of the Maxwell Demon were seriously discussed, particularly at the quantum-mechanical level [20], [21].

In the end of the 1980s - beginning of the 1990s rapid development of laser industry led to appearance of ultrafast, the so called *femtosecond* lasers. Lasers of new generations allow to generate pulses with duration about a few femto-seconds and even less (fs) ($1 fs = 10^{-15} \sec$). Duration of such a pulse is comparable with period of molecule natural oscillations. Therefore, a femtosecond laser can be, in principle, used as a mean for control of single molecules and atoms. A consequence of such an application is a possibility of realizing an alchemists' dream: changing natural course of chemical reactions.

Using apparatus of the modern control theory may open new horizons in studying interaction of atoms and molecules and find new ways and possible limits for intervention into intimate processes of the microworld. The sections 2 and 3 of the paper are devoted to application of speed-gradient method, well known in nonlinear control area [8], [11] to control of model molecular systems both in quantum-mechanical and classical setting.

2. CONTROLLING QUANTUM OBSERVABLES FOR DIATOMIC MOLECULE

Quantum model of a diatomic molecule. The following mathematical model of a controlled quantum system is considered, describing a diatomic molecule with Morse potential [22,23,24] (in Hartree units):

$$i\frac{\partial \Phi}{\partial t} = -\frac{1}{2M}\frac{\partial^2 \Phi(t,r)}{\partial r^2} + V(r)\Phi(t,r) + uAr\Phi(t,r) \quad (1)$$

$$V(r) = D(\exp(2\alpha\frac{r_0 - r}{r_0}) - 2\exp(\alpha\frac{r_0 - r}{r_0})) \quad (2)$$

where $i = \sqrt{-1}$ - imaginary unit; $\Phi(t,r)$ - wave function, for any time instant t_0: $\Phi(t_0,r) \in L^2(0,+\infty)$; r - distance between atoms of molecule, $r \in (0,+\infty)$; V(r) - Morse potential; *M, A, D*, α, r_0 - constants, which depend on specific molecule.

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A finite level approximation by eigenfunctions $\phi_k(r)$ of unperturbed Schrodinger operator: $H_0 = -\dfrac{1}{2M}\dfrac{\partial^2}{\partial r^2} + V(r)$ corresponding to undissociated molecule states is used. Eigenfunctions and eigenvalues are analytically given [22]. The finite level approximation of (1), (2) yields the model

$$i\dot{\phi}(t) = H_0\phi(t) + uH_1\phi(t), \; \phi(t) \in C^n \qquad (3)$$

where $\phi(t)$ - coefficient vector of wave function decomposition $\Phi(t, r)$ in n eigenfunctions; H_0 - diagonal matrix, element (k,k) is λ_k; H_1 - matrix corresponding to operator of control. The element (k,m) of H_1 can be calculated using the following expression: $A \displaystyle\int_0^{+\infty} r\phi_k(r)\phi_m(r)dr$.

The problem is to design the control function u(t) to obtain the convergence to the goal value Z_g of observable $Z(t) = \phi(t)^* Z\phi(t)$ of quantum system (3):

$$\lim_{t \to +\infty} \phi(t)^* Z\phi(t) = Z_g \qquad (6)$$

Control algorithm design. It is suggested to use Speed-Gradient algorithm in finite form [8,11] with the following goal function:

$$Q(\phi) = (\phi^* Z\phi - Z_g)^2 \qquad \phi \in C^n \qquad (7)$$

The convergence to zero of the goal function is equivalent to achievement of the control goal. According to speed-gradient method the control algorithm is chosen to ensure nonpositivity of the derivative of the goal function along trajectories of controlled system (3): $u = -\gamma\nabla_u \dot{Q}(\phi)$, here dot means derivative along trajectories of the system (3), ∇_u is gradient in u, $\gamma > 0$.

$$u = -\gamma\nabla_u \dot{Q}(\phi) = -\gamma\nabla_u[2(\phi^* Z\phi - Z_g)(i(H_0\phi + uH_1\phi)^* Z\phi - i\phi^* Z(H_0\phi + uH_1\phi))]$$

Assume that the observable Z commute with operator H_0. The following algorithm for controlling observables of diatomic molecule is proposed:

$$u = \gamma 2(\phi(t)^* Z\phi(t) - Z_g)(\phi(t)^* \{Z, H_1\}_\hbar \phi(t)) \quad (5)$$

where $\{\cdot,\cdot\}_\hbar$ - quantum Poisson bracket: $\{Z, H_1\}_\hbar = \dfrac{i}{\hbar}[Z, H_1] = \dfrac{i}{\hbar}(ZH_1 - H_1 Z)$.

Note that in quantum systems feedback control is unrealizable because each measurement changes the state of the system [25]. To use the algorithm the computer simulation should be performed for calculating control as the time-depended function for known initial data. In the simulation section an example of such control function for energy of HF molecule is presented.

Control algorithm analysis. Let the following assumptions hold:

1) $z_k \neq z_m$, $k \neq m$, $k,m = \overline{\{1,n\}}$, where z_k are eigenvalues of Z in ascending order;

2) $\lambda_k - \lambda_m \neq \lambda_r - \lambda_s$, $(k,m) \neq (r,s)$, $k,m,r,s = \overline{\{1,n\}}$;

3) $h_k^* H_1 h_n \neq 0$, $k,m = \overline{\{1,n\}}$, where h_k, be linearly independent eigen-vectors H_0.

The following theorem was proved giving the basis for using algorithm (5)

Theorem 1. Consider the system (3) with feedback control law (5), where Z and H_0 commute. Given the assumptions 1), 2), 3) and $z_k < Z_g < z_{k+1}$. Then for any initial condition from $M = \{\phi : z_k < \phi^* Z \phi < z_{k+1}\}$ the goal (4) is achieved.

Remark 1. Simulation demonstrated that the control law (5) asymptotically drives observable Z to the goal value from almost any initial state (except pure states).

Remark 2. The theorem can be applied not only for diatomic molecule. The result is also useful for other finite level quantum systems.

Simulation results. Algorithm (5) was applied to energy control of molecule HF, described by equation (1). Constants for HF molecule are as follows [23,24]: *M=1732, A=0.4541, D=0.2101*, $\alpha = 2.1350$, $r_0 = 1.75$ (Hartree units). Observable of energy is H_0: $E(t) = \phi(t)^* H_0 \phi(t)$. Molecular energy levels are energy of pure states. The initial value of phase vector is distributed between 1st and 2nd energy levels. In atomic units the initial energy is $- 0.1962$. The goal value for energy is -0.0484, it corresponds to 12th energy level. Gain factor γ is 100. Time for simulating is 50 femtoseconds ($1 fs = 10^{-15} s$). Fig. 1 displays energy evolution *E(t)* and control function *u(t)*, horizontal lines correspond to energy levels. The figures confirm efficiency of algorithm (5) for control of diatomic molecule observables. Also
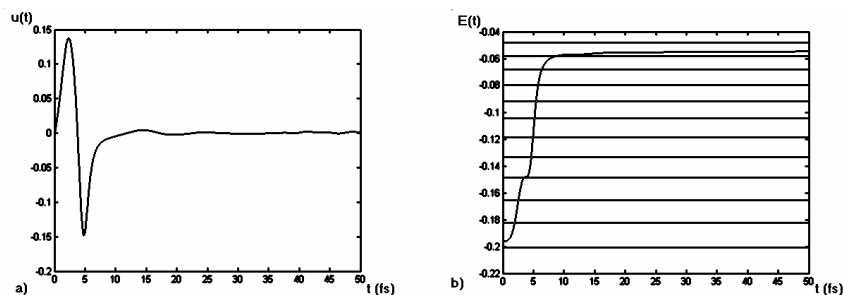
Fig. 1. **a)** control function, **b)** Energy evolution (horizontal lines are energy levels).

figures confirm that the area of stability is greater then the one for which the theorem was proved.

3. CONTROL OF ENERGY AND SELECTIVE DISSOCIATION OF A MODEL 3-ATOMIC MOLECULE

Problem of purposeful changing and stabilization of energy of internal degrees of freedom in molecular systems is an important problem of laser chemistry. Processes of the bond excitation and dissociation in weak infrared laser fields have many applications, such as control of chemical reactions, chemical structure identification and others [26]. In the final part of the paper we concentrate at the process of controlled selective dissociation of strong bond in a 2-DOF model of HCN molecule with classical mechanical representation of molecular dynamics, suggesting some new approaches to the foregoing problems.

Although molecular motion is properly described by the Schrodinger equation, successful modelling for many molecular purposes can be done with classical mechanics [27]. Thus, since the dissociation stage is characterized by the high values of energy, the results for classical and quantum-mechanical simulations of controlled dissociation are often similar (see, e.g. [28]). Often control design for quantum-mechanical ensembles is very complicated. Therefore we study the controlled selective dissociation for the model of molecule, based on classical model of the intramolecular dynamics.

The 3-atomic molecule HCN in external laser field is modelled as two kinetically and potentially coupled Morse oscillators with collinear orientation, interacting with the external field. Effects of bending, rotation and changing orientation are neglected. Despite all these simplifications, there are some fundamental difficulties in a model analysis:
1. strong nonlinearity (so the oscillations frequency depend on the energy and superposition principle doesn't realize);

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2. strong coupling between internal degrees of freedom (it leads to the additional displacement of frequencies and fast energy redistribution).

The typical behavior of the uncontrolled unimolecular system at high energy level is that deposited energy will break the weaker bond. Thus for selective dissociation of stronger bond we should control at oscillations frequency (to excite the system), and deposited energy should be concentrated in stronger bond. Additionally, control should be robust and effective when its intensity is small (arbitrarily small). The problem is complicated by the fact that control is scalar. These features makes the problem of selective dissociation nontrivial and interesting.

Model of controlled system. The full Hamiltonian of the HCN molecule in external laser field [27] is as follows: $H = H_{mol} - d(R_1,R_2) U(t)$,

where H_{mol} is molecular Hamiltonian, and the second term represents the laser-molecule interaction, $U(t)$ is the electric field amplitude (as a function of time t) – control, $d(R_1,R_2)$ – dipole moment, R_1 and R_2 are the displacements of CN and CH bond lengths from their equilibrium values $R_{1,eq}$ and $R_{2,eq}$.

Linear 3-atomic molecule is modeled as two coupled Morse oscillators. The coupling terms are of two kinds: one is a model independent kinetic coupling term and the other describes coupling within the interaction in internal coordinates. The HCN molecular Hamiltonian is $H_{mol} = H_1 + H_2 + H_{12}$ with Hamiltonians of bonds:

$$H_i = \frac{P_i^2}{2m_i} + V_i(R_i) \qquad i=1,2 \qquad (8)$$

which is composed of a kinetic energy term and a potential energy term $V_i(R_i)$ *for* bond i of the form:

$$V_i(R_i) = V_{i1} Z_i^2 + V_{i2} Z_i^3 + V_{i3} Z_i^4 \qquad i=1,2 \qquad (9)$$

where $Z_i = 1 - e^{-a_i R_i}$ $(i=1,2)$ are Morse variables, P_1 and P_2 are the conjugate momenta corresponding to R_1 and R_2.

Potential energy term (9) represents an effective potential for each bond (initially in ground, then in excited states). Expressions (8) can be called bond energies (but they are not invariants in uncontrolled system). The kinetic and potential coupling terms constitute Hamiltonian of bonds interaction:

$$H_{12} = -\frac{1}{M_C} P_1 P_2 + V_{12}(R_1,R_2)$$

where:

$$V_{12}(R_1,R_2) = V_1 Z_1 Z_2 + V_2 Z_1^2 Z_2 + V_3 Z_1 Z_2^2 \quad (11)$$

Dipole moment was chosen to simulate that of an instantaneous dipole:

$$d(R_1,R_2) = d_e (d(R_1) - d(R_2)) = d_e ((R_1 + R_{1,eq}) e^{-aR_1} - (R_2 + R_{2,eq}) e^{-aR_2})$$

Control algorithm and simulation results. For the solution of problem some methods of control theory were used, such as optimal control [27] or method of reference model on energy [24]. However, control functions, designed by these methods, have such drawbacks as nonrobustness and complicated form.

Below we apply the speed-gradient type method to the partial oscillator, corresponding to the strong bond of molecule. The natural control goal is to increase the energy of the partial oscillator corresponding to the strong bond (other bond is regarded as absent), i.e. the goal is: $Q=\gamma(H_1^* - H_1)^2 \to 0$, where H_1^* is desired value of energy (sufficiently high), γ - coefficient. According to the speed-gradient the designed control algorithm is as follows:

$$U = \gamma \dot{R}_1 , \quad \gamma < 0 \quad\quad (13)$$

$$U = \gamma \, sign(\dot{R}_1) , \quad \gamma < 0$$

Such a control looks like negative friction in strong bond and, obviously, injects energy in the strong bond. However, deposited energy can break weak bond because of energy redistribution or excitation of the strong bond. One of the possibilities to avoid this situation is to stabilize certain regimes of oscillations, providing coordinated dynamics of bonds. It is seen, that anti-phase regime provides convenient coordinated dynamics of bonds since if the system in the anti-phase regime and the sign of control is opposite to that of the strong bond velocity then excitation of strong bond and suppression of weak bond vibration take place simultaneously. It is important that such control supports anti-phase regime. I.e. if the system is initially in such regime and control is of described type then selective dissociation of strong bond takes place almost surely. A typical process is shown in Fig. 2 for system initially in the anti-phase regime and control (13) with $\gamma = -0.01$ (horizontal axis is time in units of T_0 (minimal period of small oscillations of system).

One of the main advantages of the control of such type is that synchronization of oscillations in anti-phase regime takes place for initial data from rather wide region when the control intensity belongs to a certain range.

In Fig. 3 the connection between synchronization and selective dissociation is shown. For both pictures: control function is given by (13) with $\gamma = -0.01$, initial data - $(R_1, R_2, 0, 0)$ with $R_1, R_2 \in [1.5, .. 4]$ (R_1: horizontal axis, increasing from right to left, R_2 : vertical axis, increasing from bottom to top). In

the left picture: if selective dissociation of strong bond took place (strong bond dissociates, weak bond remains unbroken), then corresponding point of plane is painted light. In the right picture: light point shows that oscillations of system with corresponding initial data are close to synchronous. If the same point on both picture is light, then selective dissociation of strong bond takes place after the synchronization in anti-phase regime.
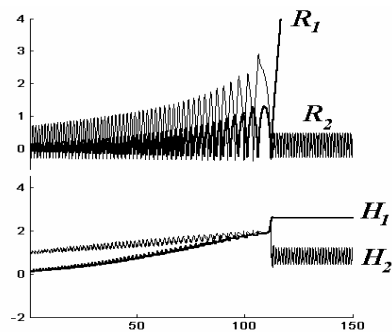

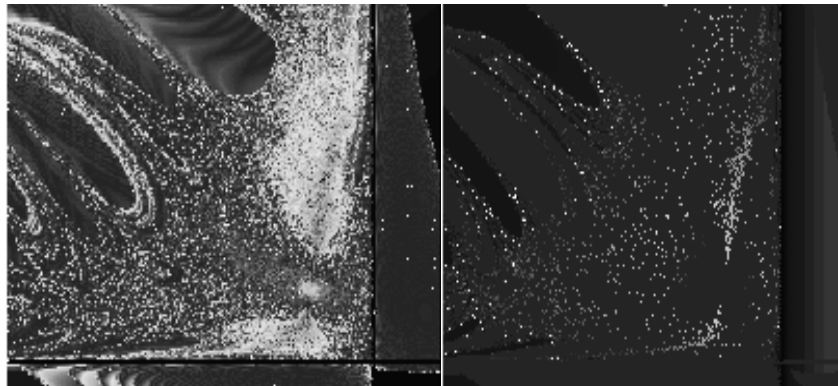**Fig. 2**: Example of selective dissociation process.

Fig. 3: Synchronization and selective dissociation.

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