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Title: Quantum control

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Quantum control

Mathematical Subject Classification

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Synonyms or related entries

control of quantum dynamics by electromagnetic radiation; laser control of chemical reactions; control of spin systems; control in Nuclear Magnetic Resonance; construction of logic gates for quantum computers

Definition

Quantum control is the control, at the quantum level, of the state or dynamical evolution of some quantum system by means of electromagnetic radiation such as a laser, a magnetic field etc. The system can be either a molecule, a set of molecules, a crystal, a protein, a spin system etc.

Overview

Controlling the evolution of molecular systems at quantum level has been considered from the very beginnings of the laser technology. However, approaches based on designing control pulses based on intuition alone did not succeed in general situations due to the very complex interactions that are at work between the laser and the molecules to be controlled, which results e.g., in the redistribution of the incoming laser energy to the whole molecule which prevents it from acting accordingly to the intuition. Even if this circumstance initially slowed down investigations in this area, the realization that this inconvenient can be recast and attacked with the tools of (optimal) control theory [5] greatly contributed to the first positive experimental results [1; 9; 17].

One regime is related to time scales of the order of the femtosecond (10^{-15}) up to picoseconds (10^{-12}) and the space scales vary from the size of one or two atoms to large polyatomic molecules.

Historically, the first applications that were envisioned were the manipulation of chemical bonds (e.g., selective dissociation) or isotopic separation. Although initially, only few atoms molecules were investigated (di-atomics) the experiments soon were designed to treat more complex situations [1]; continuing this work, further poly-atomic molecules were considered in strong fields.

But the applications of laser control do not stop here. High Harmonic Generation [2] is a technique that allows to obtain output lasers whose frequency is large integer multiples of the input pulses.

In a different framework, the manipulation of quantum states of atoms and molecules is a crucial step in the construction of quantum computers [4; 16]

A distinct, yet very related, setting is the control of spin dynamics in Nuclear Magnetic Resonance (NMR).

Finally, biologically related applications are also the object of ongoing research.

Mathematical modeling: control of the Time Dependent Schrödinger Equation (TDSE)

The evolution of an isolated single quantum system can be described by the Schrödinger equation

$$i\frac{\partial}{\partial t}\Psi(t,x) = H(t)\Psi(t,x) \tag{1}$$

starting from the initial state

$$\Psi(t_0,x) = \Psi_0(x). \tag{2}$$

where $H(t)$ is the (self-adjoint) Hamiltonian of the system and $x \in \mathbb{R}^\gamma$ is the set of internal degrees of freedom (see also in this Encyclopedia the entry "Schrödinger equation for chemistry" for additional information on this equation). We can take $H(t)$ to be a sum of a free evolution part H_0 and a part describing the coupling of the system with an e.g., laser source of intensity $\epsilon(t) \in \mathbb{R},\ t \geq 0 : H(t) = H_0 + H_I(t)$. In the dipole (i.e., first order) approximation $H_I(t)$ is written in terms of $\epsilon(t)$ and a dipole moment operator μ $H_I(t) = -\epsilon(t)\mu$. One obtains the dynamics:

$$i\frac{\partial}{\partial t}\Psi(t,x) = (H_0 - \epsilon(t)\mu)\,\Psi(t,x). \tag{3}$$

Note that higher order field dependence can also be considered $H_I(t) = \sum_k \epsilon(t)^k \mu_k$.

Beyond the situation of a single, isolated molecule, it may be interesting to study the dynamics of an ensemble of identical molecules that only differ by their initial state. The model involves the density matrix operator $\rho(t)$. The evolution equation for ρ is then:

$$i\frac{\partial}{\partial t}\rho(t) = [H(t), \rho(t)] \tag{4}$$

$$\rho(0) = \rho_0. \tag{5}$$

The density matrix formulation is also a good setting to study non-isolated systems. One way to model this circumstance is the so-called Lindblad form [10]:

$$i\frac{\partial}{\partial t}\rho(t) = [H(t), \rho(t)] + \frac{i}{2}\sum_r \left(2L_r\rho L_r^\dagger - L_r^\dagger L_r\rho - \rho L_r^\dagger L_r\right), \tag{6}$$

where L_r are operators that describe the interaction of the system with its environment. For another context isolated and non-isolated systems are described by an evolution equation involving the density matrix see the entry "Semiconductor Device Problems" of this Encyclopedia.

External fields can also be used to manipulate molecules to achieve molecular axis alignment or orientation, cf. [15] and references therein.

In Nuclear Magnetic Resonance (NMR) the control operates on the spin variable (and not on the spacial part of the wavefunction). The basic set-up in NMR consists of an ensemble of N spin-$\frac{1}{2}$ particles (e.g. electrons) subjected to a magnetic field. The evolution of the system can be written as above with the distinction that H_0 may be null and the only nontrivial part of $H(t)$ is the coupling $H(t) = \sum_k \omega_k(t)\mu_k$ with the magnetic field; here $\omega_k(t)$ are controls. Each particle lives in a 2-dimensional Hilbert space (one dimension for each value of the spin) thus the system lives in a 2^N-dimensional (direct product) space.

Controllability

A first important question is whether is it possible to control the system to a desired prescribed final state or to set a certain property or measurement to a desired value. If for any compatible couple of initial and final states a control $\epsilon(t)$ exists such that a system starting from the initial state reaches the final state by the final time then the system together with its interaction is called controllable. General tools of controlla-

bility in Lie groups can be applied (cf. [3; 12]) which allows to obtain controllability criteria such as:

Theorem 1. *If the Lie algebra $L_{-iH_0,-i\mu}$ generated by $-iH_0$ and $-i\mu$ has dimension N^2 (as a vector space over the real numbers) then the system (4) is density matrix controllable (which implies that (1) is also controllable). Furthermore, if both $-iH_0$ and $-i\mu$ are traceless then a sufficient condition for the density matrix (thus wavefunction) controllability of quantum system is that the Lie algebra $L_{-iH_0,-i\mu}$ has dimension N^2-1.*

Another set of results [11] gives sufficient conditions in terms of the so-called *connectivity graph* and of the spectrum of H_0.

Finally, one may ask what happens when several identical molecules (differing by their orientation with respect to the incident beam) are submitted to the same control. It can be shown that if any member of the ensemble is controllable then the entire ensemble should be controllable. This very strong positive result is rather counter-intuitive and it arises as a result of the non-linearity of quantum control.

Finally for infinite dimensional controllability encouraging results obtained using tools in nonlinear control have already been obtained by K. Beauchard, J.M. Coron, V. Nersesyan, etc.

Optimal control

Construction of the cost functional

Assessing the controllability of a system does not necessarily imply that a constructive mean to find a convenient control is available. Especially for complex systems, in practice it is necessary to use experimental or numerical procedures to find the control. One approach that can be used to treat this situation is the optimal control theory which is based on the introduction of a *cost functional* (also named "quality index"

or "quality functional") depending on the driving controlling field that describes the target, additional costs and whose optimization gives an convenient field.

A simple example of a cost functional is the additive form where it depends only on the final state $\Psi(T)$ and the laser characteristics

$$J(\epsilon) = \langle \Psi(T)|O|\Psi(T)\rangle - \alpha \int_0^T \epsilon^2(t)dt, \tag{7}$$

where $\alpha > 0$ is a parameter and O is the observable operator that describes the goal: a large value $\langle \Psi(T)|O|\Psi(T)\rangle$ means that the control objectives have been conveniently attained. Recall that (for a single system of wavefunction $\Psi(T)$) $\langle \Psi(T)|O|\Psi(T)\rangle$ can in practice be computed as an average over experiments corresponding to measuring the observable operator O. Examples of observables O include the projection to a predefined target state Ψ_T, spatial depending functions $O(x)$ etc. See also in this Encyclopedia the entry "Schrödinger equation for chemistry" for additional examples of observables.

When the system is represented through a density matrix $\rho(t)$ measuring the observable O allows to compute $Tr(\rho(T)O$ and thus the natural cost functional is:

$$J_d(\epsilon) = Re(Tr(\rho(T)O)) - \alpha \int_0^T \epsilon^2(t)dt. \tag{8}$$

Of course, many other functional types can be constructed.

Optimization of the cost functional

In order to optimize such a functional one may be tempted to use the Pontryagin maximum principle which give the first order necessary optimality conditions [6]. However in practice different procedures, the so called *monotonically convergent algorithms*, were found to be better fitted to solve these equations. These algorithms have the very convenient property to improve the cost functional J at each iteration. In the Zhu & Rabitz formulation [11] the iterations indexed by $k = 1, 2, ...$ are carried on following the formulas:

$$\begin{cases} i\frac{\partial}{\partial t}\Psi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\Psi^k(x,t) \\ \Psi^k(x,t=0) = \Psi_0(x) \end{cases} \tag{9}$$

$$\epsilon^k(t) = -\frac{1}{\alpha}\mathrm{Im}\langle\chi^{k-1}|\mu|\Psi^k\rangle(t) \tag{10}$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x,t) = (H_0 - \tilde{\epsilon}^k(t)\mu)\chi^k(x,t) \\ \chi^k(x,t=T) = O\Psi^k(x,T) \end{cases} \tag{11}$$

$$\tilde{\epsilon}^k(t) = -\frac{1}{\alpha}\mathrm{Im}\langle\chi^k|\mu|\Psi^k\rangle(t) \tag{12}$$

An important property of this algorithm is that if O is a self-adjoint positive semi-definite observable then the algorithm converges monotonically in the sense that $J(\epsilon^{k+1}) \geq J(\epsilon^k)$.

More general formulations (including the density matrix versions and also open systems) are to be found in [14]; the even more abstract approach of [13] identifies what is the most general setting where a monotonic algorithm will work and gives the formulation of the algorithm. The methodology in [13] works not only for nonlinear Hamiltonians but also for multiple coupling fields.

Stabilization by Lyapunov functionals

The quantum tracking procedures (e.g. [11]) also called local control procedures obtain explicitly the control field from the prescribed trajectory that the system is required to take. Such methods are appealing numerically since it is expected that they only requires a few propagations of the Time Dependent Schrödinger Equation (TDSE).

Introduce the performance index $y(t)$ that formulate the desired physical properties to be satisfied by the system, defined as $y(t) = y(\langle\tilde{O}_1(t)\rangle, \langle\tilde{O}_2(t)\rangle, ..., \langle\tilde{O}_N(t)\rangle)$. Here $\langle\tilde{O}_j(t)\rangle$ for $j \in \{1, 2, .., N\}$ denote the expectation value of the physical observables equal to $\langle\Psi(t)|\tilde{O}_j(t)|\Psi(t)\rangle$ or $Tr(\rho(t)\tilde{O}_j(t))$; these (Hermitian) observables $\tilde{O}_j(t)$

are supposed to follow the dynamics $i\frac{\partial}{\partial t}\tilde{O}_j(t) = [H_0, \tilde{O}_j(t)]$ A simple computation shows that:

$$\frac{dy(t)}{dt} = -\epsilon(t) \sum_{i=1}^{N} \frac{\partial y(t)}{\partial \langle \tilde{O}_j(t) \rangle} \langle [\tilde{O}_j(t), \mu/i] \rangle. \tag{13}$$

In particular the feedback

$$\epsilon(t) = -\sum_{i=1}^{N} \frac{\partial y(t)}{\partial \langle \tilde{O}_j(t) \rangle} \langle [\tilde{O}_j(t), \mu/i] \rangle \tag{14}$$

ensures $dy(t)/dt \geq 0$. La Salle theorem and variants (see [7] Chap. 4.2) is used to derive convergence results (see op. cited) for such algorithms.

Experimental and stochastic algorithms

Laboratory realization of quantum control experiments builds on the coupling between the experimental apparatus with convenient optimization algorithms that search within the set of control fields the optimal individual. In the experimental setting a zero order optimization algorithm (i.e. that only uses the value $J(\epsilon)$ of the functional and does not need its derivatives) is run on a computer [11]. Each time that this algorithm requires to evaluate J for a candidate field ϵ the field is created and the outcome measured and handed over to the optimization algorithm.

Of course, a numerical version of the algorithm can be used too where a numerical procedure is used instead of an experiment in order to create the field and compute the cost functional.

It is important to mention that this procedure is enabled by the very high experimental repetition rate available (as many as a thousand shots a second).

In practice Genetic Algorithms (GA) have been used and latter superseded by Evolutionary Strategies (ES). Both procedures can be formally described as following the steps: selection of the parents that will generate offsprings based on the fitness

of the individuals; application of the evolution operators such as mutation and cross-over; evaluation of the fitness of offsprings; replacement of the current generation by a new one according to specific criteria that e.g. can allow the parents to survive or not; evaluate the stopping criteria and if these are not met then move to the next generation [11].

Inverse problems and other applications

The ability to generate a large amount of quantum experiments and measure the results may be exploited as a possibility to learn more about unknown parameters of the quantum system itself. From the mathematical point of view we enter the field of the "inverse problems" where some parameter characterizing the system is found from measurements; it has been formulated within an optimization framework in various settings [11; 8].

Two types of questions are usually relevant to this topic: first a theoretical question concerns the well-posedness what can be said about the existence and the uniqueness of the Hamiltonian, and/or the dipole moment, etc., compatible with a given set of measurements; secondly what are the best algorithms to recover the unknown parameters from measurements. We refer to the cited works for details.

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